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BULLETIN OF THE RESEARCH COUNCIL OF ISRAEL

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Papers dealing with scientific problems of regional interest will be considered for publication. Intending contributors, in preparing papers for submission, should follow the general style of this Bulletin.

Papers, which should be written as concisely as possible, should be typed on one side of the paper only, double-spaced, with side margins at least 2.5 cms. wide. Pages, including those containing illustrations, should be numbered.

Summary

A short summary should precede the paper.

Tables

These should be labelled and numbered in Roman numerals.

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Line drawings should be in India ink on white drawing paper, bristol board, tracing paper, blue linen or blue ruled graph paper. They should be clearly drawn and be capable of reduction to 17×13 cms, without loss of detail. Lettering should be inscribed in soft pencil.

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3) Taylor, G.I., 1932, Proc. R. Soc. (London) A, 138, 41.

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This Bulletin, which is published by the Research Council of Israel, has a two-fold purpose. Primarily it is intended to facilitate the publication of scientific papers connected with problems of regional import but of wider interest. In addition, preliminary communications on the result of both fundamental and applied research will be accepted in the form of Letters to the Editor.

Thus, the Bulletin will record the results of research carried out in the scientific institutions of Israel or directly sponsored by the Research Council and which have previously appeared scattered in scientific journals all over the world.

The first Bulletin appears as a double number, and it is intended in the future to publish four issues annually.

The contents of this publication were all received before 1 September 1950, when this Bulletin was ready for the printer. Due to the temporary lack of technical facilities, however, publication has had to be delayed till the present date and certain makeshifts have had to be employed.

Executive Editor.

1 March, 1951.

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This paper is dedicated to the memory of G.S. Blake, pioneer in the geological exploration of this country.

GEOMORPHOGENY OF ISRAEL

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PART I — THE NEGEV

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III Sedimentary Southern Negev

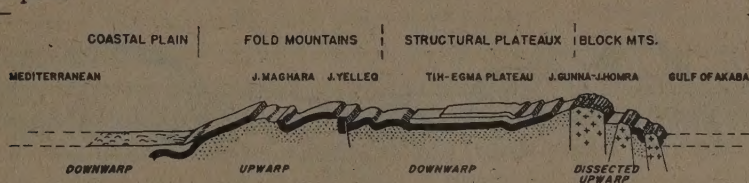
Introduction

The morphology of Israel is an expression of its tectonic structure. The harmony between its relief and structure is most clearly demonstrated in the Negev and in Galilee. In an unusually plastic manner, the dominating block-structure of Galilee and the impressive fold-structure of the Negev are revealed by the form of the mountains and the valleys, and sometimes, as in the Negev, directly by the hydrographic pattern. The sculpture of the landscape is emphasized in the Negev by the arid nature of the climate. There is no trace of the Caliche-crust known as Nari. Even in the foothills sloping down to the west and in the uplands there is none of the Mediterranean terra rossa which predominates in the north. The mountainous Negev is naked and devoid of all protective soils. The layers of dust and sand which have been preserved as secondary products in the lowlands and more rarely on certain plateaux, are often mingled with mud and gravel sediments deposited by torrents in different epochs. The relief effected by running water still makes itself felt in short seasonal floods, whose force, however, lags far behind the hydraulic abrasive power of the earlier Neogene and particularly of the Pleistocene-Pluvial rivers. In the central Negev, and even more in the southern area the wind plays an increasingly deflating role : there are ventifacts in the southern Araba plain.

Between the Israel and the Sinaitic or Egyptian Negev there are no climatic or morphological boundaries, and the present boundary is also artificial from the structural aspect. In the north-western corner of the Sinai peninsula the dune dominated coastal plain runs into the Gaza plain. In the north-eastern corner of the Gulf of Akaba the Precambrian or crystalline Sinai is continued a few kilometres over the boundary at Elath. The rift-valley border between Wadi Meneiaie and Wadi Masri, strongly faulted and split up into small blocks, constitutes a province of its own. From the morpho-tectonic point of view this area is best ascribed to the western wing of the crystalline-framed Akaba-Graben. It is, therefore, not included in the following investigation.

The *southern Negev*, which in the main includes the intake area of the Wadi Jerafi, drains a section of the Tih-Egma plateau, which is otherwise subordinate to the Wadi system of Arish. The *central Negev*, which includes the area of the culminating anticlines of Wadi Raman and Jebel Ureif, is continued into the Sinai peninsula at the zig-zag Egypto-Israel boundary, where the dome of the Araif-en-Naja attains the height of 934 metres. The central Negev thereby becomes the central structural upwarp of the Negev, which dips into downwarps from this point on all sides. Radial drainage of the Wadi Hafir, Quseib-Merzaba, Jerafi-Heiyani and Gerai-Arish is a characteristic feature of these *regions of downwarps*. The main morphologic elements of the downwarp region are cuesta — and mesa — forming plateaux; where folds occur, they are low and small. The crystalline Sinai appears thus as a gigantic *upwarp massif* split up into blocks, joined in the north to the structural plateaux of Tih-Egma with its giant cuestas and the *downwarp of Nekhl* (Wadi El-Arish, el-Bruk, el-Gerai). The anticlines following on the Nekhl downwarp culminate in the Jebel Maghara and Jebel Jelleq (+1090m.). They assume the same tectonic and orographic as well as morphologic position as the central Negev of the Raman-Ureif-Araif-en-Naja. The *Maghara-Yelleq upwarp* dips below the coastal plain at el-Arish — Rafa. Our Negev coast together with the downwarp of Auja-el-Hafir — Khalasa forms a regional major depression, a main reservoir of the youngest *continental sediments*: inland-dunes and adobé-loess. In the accompanying block diagram (fig.2) (hitherto unpublished, though the author has made use of it for university lectures during the past ten years) our morpho-tectonic conception of the Sinai is reproduced. It will be seen from this diagram that as regards structure and relief, the Sinai forms the counterpart — in a larger dimension — of the central Negev. The *north Negev* following on in the north-east betrays still smaller dimensions (fig. 5): the elevated anticlinal ridges are grouped round the *Kurnub (Wadi Hathira) upwarp* (+700m). The *Khalasa and Sab' — Milh downwarp* (dendritic Wadi pattern) of the west belongs to the giant depression of the coastal region. The eastern downwarp coincides with the rift-valley region of the El-Ghor — Dead Sea depression. The fold-morphology is most pronounced in the vicinity of the upwarp region of Kurnub. It fades away to the east in the direction of the

graben and to the west in the low foothills of Khalasa — Beersheba and the Gaza coastal plain.



BLOCKDIAGRAM—SECTION THROUGH SINAI

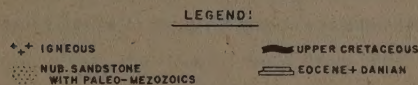


Fig. 2.

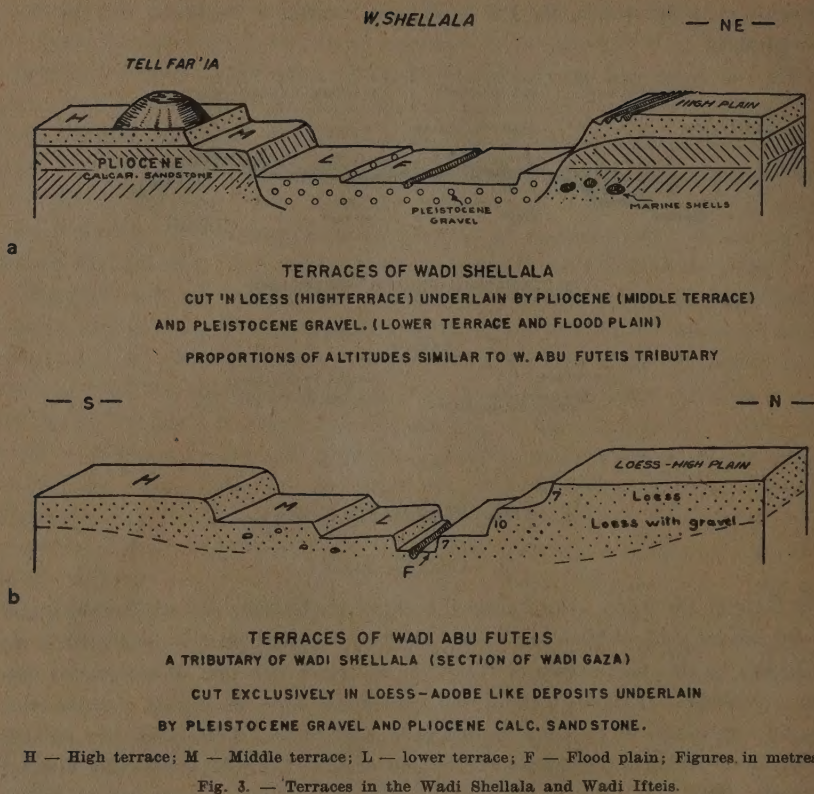
I. THE NORTHERN NEGEV

1. COASTAL PLAIN

The Gaza or the *Negev coastal plain* is both morphologically and structurally a part of the coastal plain of Sinai and Israel. The Negev coastal plain is dominated in the first place by the main stream of Wadi Ghazza, which falls into three sections: Shiniq — Shallala — Ghazza. Its transversal consequent course harmonizes with the other coastal consequents of the Israel coastal plain. Longitudinal subsequents join up with it only in the region of the north-south situated locations Kh. el Main—Beeroth Yitzchak—Bureir. Thus there is formed a remarkable rectangular pattern, peculiar to this region, which clearly reflects the substructure of the so-called Huleiqat dome, and also demonstrates the very recent relief of the coastal plain.

The very recent origin of the coastal wadis is also demonstrated in the constant middle-terrace of Wadi Ghazza which is remarkably well preserved, despite the extremely loose matter of the so-called loess in which its course is incised. The coastal section Ghazza and Shallala and other tributaries of the main drainage channel are entirely embedded in the layers of the partly fluvialite deposited or redeposited adobé-like loess. Plio-Pleistocene outcrops appear only in the river bed of the middle-course at Ein-Shallala (fig. 3).

The evolution of the middle-lower course of the Wadi Ghazza is, at best, Upper Pleistocene and that of its tributaries of the Holocene period. It is, therefore, not surprising that there are few main tributaries and that a coarse branching pattern only appears at the approach of the foothills.



2. FOOTHILLS

The transition of the Israel coastal plain into foothills of the Shephela region is marked by a fall-line situated at a height between + 100m and + 150m. In the Negev the fall-line is covered by loess and moving inland dunes (Shaluf of Khalasa): the step-like western edge of the foothills, familiar from the Shephela, is thus hidden. Possibly the outcrops of the Pliocene beach-conglomerate to be found in the bed of the Wadi at Bir-Shiniq, Bir Ifteis and Tell esh Sharia represent the postulated fall-line. In an earlier paper (1936) we ascribed a more structural significance to this line (structural line I). In any case the Pliocene outcrops of Wadi Shiniq mark the beginning of a general incision of the rivers into the chalk and flint-chalk series of the Eo-Oligocene mountain formation, and hence a branching of the Ghazza master-stream into three main upper courses: Sab', Sidd and Khalasa. Each of these upper courses branches out

again into further subsidiary tributaries. The *coarse dendritic Wadi pattern* characteristic of the Shephela foothill region, wherein the main branches run in consequent and transversal direction to the coast, is also to be found in the Negev foothills. Thus the pattern of the foothills in no way reveals the folded subsoil. Even at the approach to the uplands, where the structure of the folds stands out in relief as in the anticlinal ridge of Jebel-esh-Shereif or in the synclinal valley of Asluj—Subeita—the drainage is always through the transversal consequents. Jebel Qarn constitutes the only exception with its longitudinal strike Wadi Nehiya. A definite change of the dendritic pattern into the trellis-pattern occurs only at the + 400 m contour line of Auja-el-Hafir—Asluj-Biyar Arara with the appearance of the anticlinal ridges of Meqrah—Beqqar. Here end the foothills, and the fold uplands begin, and, here, in contrast to the foothills, structure and relief converge. This raises the question whether the morphology of the foothills is possibly young like that of the coastal plain, so that erosion has not yet scooped out the fold structures. Alternately it may be much older than the “premature” uplands. In any case the Holocene — Upper Pleistocene loess and inland dunes cover the old drainage channels, as for example those of Wadi Ruheiba, southwest of Khalasa and also, as we have seen, the fall-line. The foothill-wadis are, therefore, older than the coastal consequents. This is still more clearly demonstrated in the longitudinal and transversal cross sections of the wadis. The main channels of the Arara, Sab', Shugheib, Sidd, Khalasa and Ruheiba run through the very wide basin-shaped valleys banked by gravel terraces forming broad, meandering loops. Innumerable little minor valleys with insignificant wadis and even gullies drain into these main valleys. As a result, the main channels are poor in real tributaries. The drainage of the foothills is carried out principally by the main channels themselves, thus determining the coarse dendritic character of the wadi pattern. They are very old inherited river-courses which in the rainy season act as broad directive water-roads in the labyrinth of the low, well-rounded chalk hills.

These old transversal foothill consequents, like the present-day coastal consequents, must date back to an earlier coastal plain or to some other *initial surface*, that was inclined to the West. In fact there exists that remarkable surface abruptly cutting off the folds of the foothills, which we recognized and defined (1943)* as the marine abrasion plain of the Vindobonian (stage a, fig. 4) sea. This Miocene initial surface, raised and tilted in the Pontian, became the coastal plain of the succeeding Asti-Plaisancian sea. Upon this *Mio-Pliocene coastal plain* there developed the transversal consequents of the *first or Mio-Pliocene main cycle* of erosion (stage b, fig. 4). They entered the Pliocene sea in the region of the fall-line postulated above (fig. 4).

The upwarp movement at the end of the Pliocene (Levantian) induced a new

* PICARD, L., and SOLOMONICA, P., 1936, The Geology of the Gaza-Beersheba Plain, Bull. Geol. Dep. Hebrew Univ. 1, (Jerusalem).

rise and therewith an enlargement of the Pliocene coastal plain, and gradually the development of the *Pleistocene coastal plain*. The rivers which then penetrated the foothill "hinterland" with its tilted peneplain attaining a height up to 310 m cut themselves deeper into the soft chalk rocks, but also broadened their profile into "pseudoglacial" valleys. (In the hard rocks of the Judean hills well developed valley-shoulders mark the beginning of this new stage). The *second or Plio-Pleistocene*

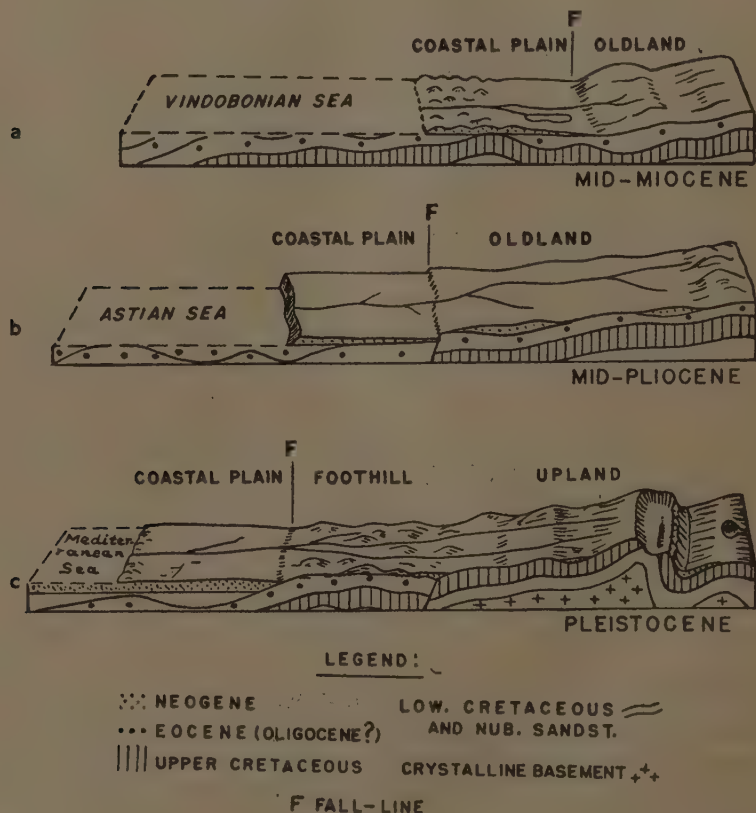


Fig. 4. — Evolution of the Western Negev and Coastal plain.

main cycle of erosion then occurred, attaining a climax in the main Pluvial or the old Pleistocene. The rivers of the foothills at this period discharge their material into the Pleistocene coastal plain near the fall-line and in building up gigantic alluvial gravel fans produce a piedmont slope, extending almost to the coast (stage c, fig. 4). Slight mid-Diluvial subsidence of the coastal plain (together with a slight marine ingression) accounted for the blanketing of the young Quaternary layers of loess and dunes in the south, of Kurkar, Hamra and red sand in the north. The upper Pleistocene-Holocene coastal drainage has so far only succeeded in incising to the Piedmont gravel, and here only in the main wadis of the lower course. Contemporaneously, the flood plains developed in the main wadis of the foothills.

3. THE NORTHERN UPLAND

a. General

Whereas the gradient of the foothill — peneplain (250 m on 35 km) and of the coastal plain (150 m on 35 km) is gentle, the uplands rise more rapidly to the watershed (fig. 4 and 5). In one section of 18 km between the Asluj-Subeita plain (400 m) and the summit of the Kurnub upwarp (700 m) 300 m of gradient are surmounted. The same proportions apply to the central Negev upland. Here on a distance of 35 km there occurs a rise of 600 m up to the watershed of the Raman upwarp (fig. 7).

The relief changes and becomes increasingly pronounced as the steep rise proceeds. The hard Cretaceous rocks, hidden in the foothills by a cover of Eocene, now appear on the surface bared of this covering. The thick flints of the Maestrichtian, frequently also the hard limestone and dolomite of the Turonian and Cenomanian form the capping rock of the anticlinal ridges. These, however, have long been affected by atmospheric agencies. A number of short ruzes, of long transversal wadis, of cluses and gaps, of erosion cirques with and without longitudinal subsequents produce a classical fold relief and yield a typical fold-drainage pattern with the synclinal consequents. In various stages of erosion which do not, however, pass the premature stage, a genuine *Jura-morphology*, with characteristic but coarse-textured, *trellis-pattern* develops in the northern upland from Arara to Hosb and from Subeita to Zuweira. No mature inversion relief or finely nerved pattern, as in the Appalachians is however, attained either in this northern or in the more pronouncedly sculptured central and southern Negev upland. In none of the other uplands of Israel is such a "perfect" adjustment achieved between inner structure and outer sculpture as in the northern Negev. In this respect it surpasses its model — the Jura mountains.

The following somewhat schematic cross from the Mediterranean over Kurnub summit to Hosb (fig. 5) may serve to demonstrate the morpho-genetic processes.

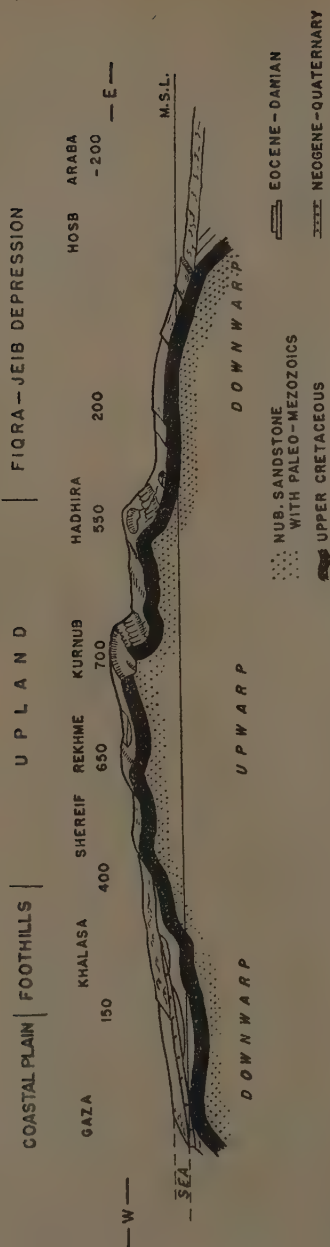


Fig. 5 on this page gives a clear idea of the anti-and synclinal structure and the pronounced eastward asymmetry of the folds. By contrast with the synclinal character of the foothill region and the Araba-graben, the Kurnub upland appears as a folded upwarp. It is approximately 25 km wide and consists of the central or *culminating anticline of Kurnub* round which one or two anticlines are grouped east and west respectively. Bare and devoid of even slight sediment cover, each anticline forms a fold-ridge, each syncline a fold-valley. In the synclinal valleys the soft marls and the chalk of the Danian-Paleocene and Lower Eocene are well developed; the interchanging layers of chalk, limestone and flint of the Middle Eocene are, however, greatly reduced by erosion. When preserved they form remarkable relic-mountains or buttes, called "Madra" by the Beduins. On these synclinal sediments of Danian-Eocene age there rest both east and west of the Kurnub saddle the Neogene inland sediments of the Hosb series.

Fig. 5. — Block diagram section through Northern Negev.

b. *Western Part*

The discovery of Miocene giant oysters (*Ostrea gignesis*) in the Wadi Rakhma at 500 m height, becomes a significant factor in our present morphogenic analysis. Like the Miocene outcrops of Kh. Khuweilifa and Kh. Murran north of Beersheba, they prove the extension of the Miocene coastline up to the region of the present Uplands, at Kurnub approximately up to the watershed. They justify, therefore, the assumption that the abrasion plain of the Miocene must have extended from the foothill region almost to the watershed. This means that roughly as far as Kurnub the folds were truncated and covered with a thin marine Miocene sediment layer. Soon the transversal coastal consequents cut their beds in the raised (Pliocene) coastal "Pene" plain. Thus the Pontian and Pliocene rivers transversed not only the above-mentioned region of the foothills, but also the western upland at least as far as the neighbourhood of the present-day watershed.

This explains the superimposed character of the steep-walled wadis called Daiqa or Daiyqa. With wide curving — sometimes incised — meanders, they breach the anticlines of Meqrah, Haleiqim, Beqqar, Rakhma, entering the Asluj — Subeita plain with characteristic "dry" gaps. The epigenetic transversal wadis also include the incision of Wadi Sirr near et Thulma. North of Kasr es Sirr, the Wadi Sirr crosses the Ras Wadi Juraba or Magrun saddle.

The transversal valleys are thus *inherited* water arteries of the first or Neogene erosion cycle. It was only after the end-Pliocene upwarping, and as a result of old-Pleistocene-Pluvial erosion, that they developed their deep gorge-like incisions and convex slopes. In this second main cycle, the raised truncated anticlines of the upland are cleared of the Neogene and of the Danian-Eocene weak cover; their summits are carved out down to the Cenomanian and their flanks worn down to the flint capping rock.

In the horizontal or gently dipping strata of the synclines, the *longitudinal wadis* following the strike are formed during the Pleistocene cycle. They cut but slightly downward into the sediment — their erosion is lateral. Their course is relatively short and straight and terminates at right angles into the older epigenetic transversal valleys. The longitudinal channels are, therefore, major consequents of the Pleistocene drainage. They are only subsequent in regard to the old transversal consequents of the Pliocene. The rectangular pattern thus formed is further constituted by *secondary lateral consequents*, which drain the flanks of the anticlines and flow into the major longitudinals. Because of the strong eastward asymmetry of the saddles, the lateral consequents are short on the eastern flank and long on the western flank (e.g., Kurnub and Zuweira anticline). Correspondingly the more symmetrical anticlines (el Meqrah) of the lower West (and of the foothills) are distinguished on their flanks by lateral consequents of more or less equal length. More dome-shaped structures such as the Shejerat el Beqqar display a regular centrifugal radial pattern. Thus in the western

region of the uplands we find a varying network of wadis which, together with the main transversals, always form a *trellis-pattern*.

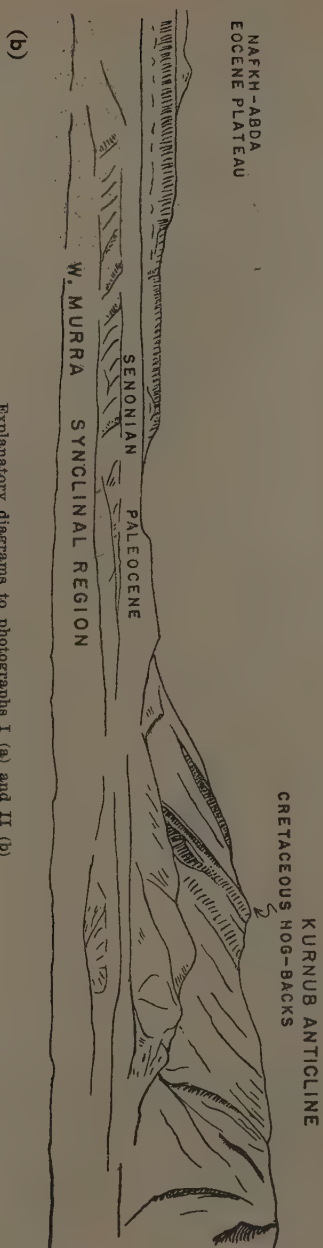
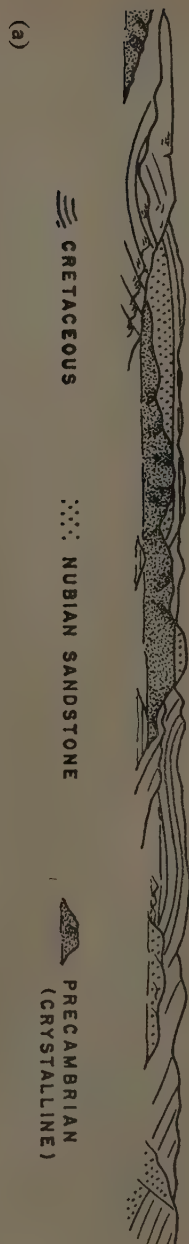
By comparison with the culminating Kurnub anticline and the other folds in the eastern section of the northern Negev, the evolution of the trellis-pattern of the western uplands is less advanced. In the western anticlines, for instance, the secondary laterals of both flanks are much less numerous and much less deeply incised. Hence there is no union yet between these laterals and hence there are no secondary transverse valleys. In the western section we only meet with primary epigenetic master streams. There the crescent-shaped cluses and the Hadhira cirques of the east are also lacking. Even their "embryo" origins, — the ruzes, are lacking. In view of the heavy western rains and the far more arid nature of the east, we cannot attribute to the climate the less accentuated erosion in the west. The cause lies structurally in the much lower position of the eastern erosion bases. The eastern *general* basis lies 200—300 m deeper because of the Araba — Dead Sea trough. (In the Pluvial period it was probably deeper still). Further, the much more pronounced asymmetry of the eastern anticlines induces the development of much more steeply inclined, shorter eastern flanks — and hence leads on the eastern foot to deeper seated *local* erosion bases. The more forceful westward headwater erosion of the northern Negev is a result of the local as well as of the general east-asymmetry of its structure. This consistently brings about a westward shift of the main and subsiding *watersheds*. The *younger* or Pleistocene Araba system of the wadis, Fiqra, Muhawat and Saiyal, therefore, dominates over the older or Neogene Ghazza system.

c. *Watershed region*

After the end-Pliocene *upwarp*, the watershed was situated on the structural summit of the Kurnub anticline, i.e., on the asymmetrical flexure axis approximately on the side of the subsequent Wadi Madsus of the easternmost portion of the Hathira funnel. With the Pluvial erosion and development of the giant erosion crater, the watershed shifted further and further westward and has today reached the western edge of the crêts of the erosion "ellipse". Hence on Jebel Hathira the shifting of the watershed totalled only 3-4 km westward during the Quaternary. The amount of shifting in the neighbouring region of the Raman upwarp in the central Negev is much greater; thus the original watershed moved 25km westward from the south crest of the Raman cirque to the original region of Wadi Murra (near Abda). In consequence the structural plateaux of Nefkh and Abda, which will be discussed later, are no longer tributary to the Auja el-Hafir system. Their former adherence to the foothill drainage of the Hafir and Khalasa — Ghazza system is demonstrated only by the consequent transverse and coarse dendritic net of the north-west draining Nefkh system. Today Wadi Nefkh with its branches, the Wadi Miqra, Abu Suna, Ramliye makes a sharp bend

eastward. Near Ein el Mureifj-Ein el Murra the Wadi Nefkh drops in a steep and narrow tapping-gorge (cut in the Middle Eocene Abda limestone), and continues into Wadi Murra — Madra — Fiqra, which drains eastwards. The Nefkh, which flowed into the sea near the Sheneq fall-line in the first cycle during the Pliocene by way of Wadi Beqera — Theiqat el Amirin — Thamila and Khalasa, was decapitated in the Pleistocene by the strong east-system of the Fiqra — Jaib and was diverted with a sharp elbow of capture into the Murra Valley. Structurally the region of capture coincides with the plunging of the Kurnub *upwarf* into the Auja el-Hafir downwarf. The Abda region is therefore a typical example of river piracy. It is distinguished not only by its tapping gorges but also by its several small upland plains without outlet. Several terrains of such "dead" drainage are to be found from the El Metrada plateau to the plains at the passes of Naqb el Ghareb and Naqb Imratama. Their character of natural aerodomes may become of no less importance than the rôle they played hitherto in camel transport. These almost non-dissected pre-Diluvial plains at 500 m altitude may well have concealed continental (Hobs-) or marine Neogene formations under their alluvial dust-and playa sediments. All the lateral wadis diverted to the Wadi Murra and departing from the high plateau of Nefkh (from Abda to Naqb-el-Hemle) have deep entrenched gorges, often with erosion-canyons many kilometres in length. The most important, the Wadi Umm Ke'ab and Mesheishe esh Sherqiye, cut into the quasi horizontal structural rock benches by headwater erosion. On the heights of the south-western branches of the Murra tributaries there is an esplanade landscape rich in cliffs — a second type of structural plateau morphology. Capture of the upper wadis by the *east system* may also account for the elbow and gorge of Wadi Kurnub-Yerqa at the Kurnub Police Post. Much more apparent, however, is the shifting of the watershed at the northernmost end of the Negev uplands in the region of the anticline of Ras-Zuweira, which plunges into the Judean desert. There the headwaters of Wadi Milh were attached to the impressive transverse gorge of Wadis Muhawwat and Saiyal. This gave rise to the poorly drained upland plain of 500 m altitude at Tell-Urad (see map fig. 1).

The hydrographic-tectonic position of the Sab'-Milh drainage can, therefore, be compared in its basic features with the Auja el Hafir drainage. In the south there lies, between Kurnub and the Raman *upwarf*, the structural basin of Hafir with consequent coarse dendritic water arteries which have in part become tributary to the Araba—Dea Sea system. In the north at Zuweira, between the plunging spurs of the Kurnub — and the Hebron *upwarf*, there appears the dendritic headwater network of the synclinal depression of Beersheba—Tell Milh, which continues into the Judean desert.



Explanatory diagrams to photographs I (a) and II (b)

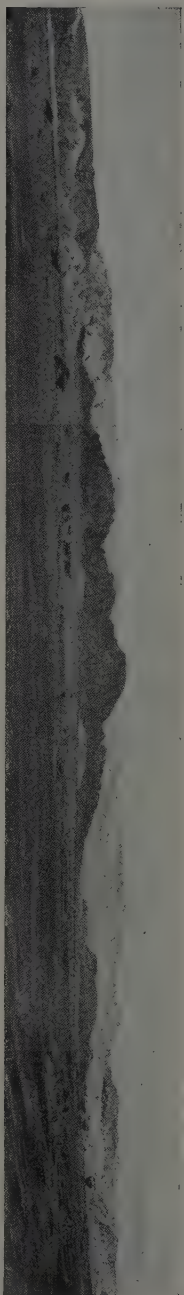


Photo I

Half-cirque of Jebel Hindis on the border of the Araba rift-valley

(Photo by S. Shitran)

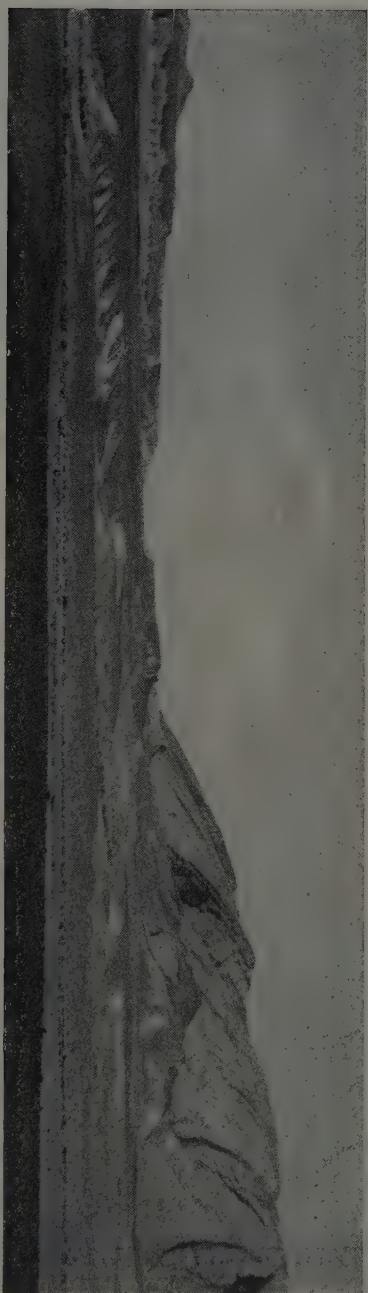


Photo II

Kurnub SE-flank with view to Abda Plateau

(Photo by F. W. Fairbridge)



(Photo by E. Locker)

Raman erosion funnel

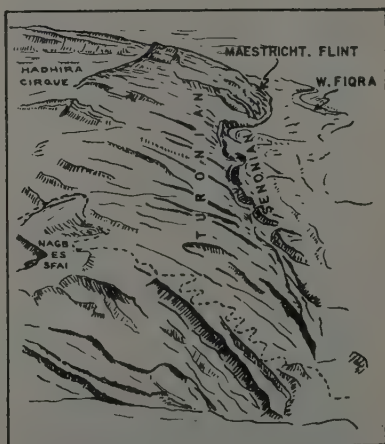
Photo III.



Photo IV.

N-E View to Nagb es Sfai and Hadhira

(Photo by courtesy of Keren Kayemeth)



N-E VIEW TO NAGB ES SFAI AND HADHIRA

Explanatory diagram to Photo IV

d. *Eastern Part*

The anticlinal ridges of Kurnub and Zuweira with their west flanks, on the one hand, still belong to the western Negev. Their main drainage, on the other hand, is through the eastern wadi system. The anticline of Jebel el *Hadhira*, however, belongs completely to the eastern desert. Here we have a classical example of fold morphology. Further eastward we have one or two shallow anticlinal ridges with but slightly pronounced relief, forming the dipping end of the Raman *upwarp*. Their low-lying folds are often covered with Neogene Hosb-series and Pleistocene Wadi Jaib-gravels. They no longer belong to the uplands but to the *eastern-lowland* — the Jaib—Fiqra depression. Some of the north-east — south-west valleys may well date back to the Neogene cycle, although their present formation mainly displays the influence of the Pluvial — Quaternary erosion. The apparently uni-cyclic erosive effect is displayed still more in the relief.

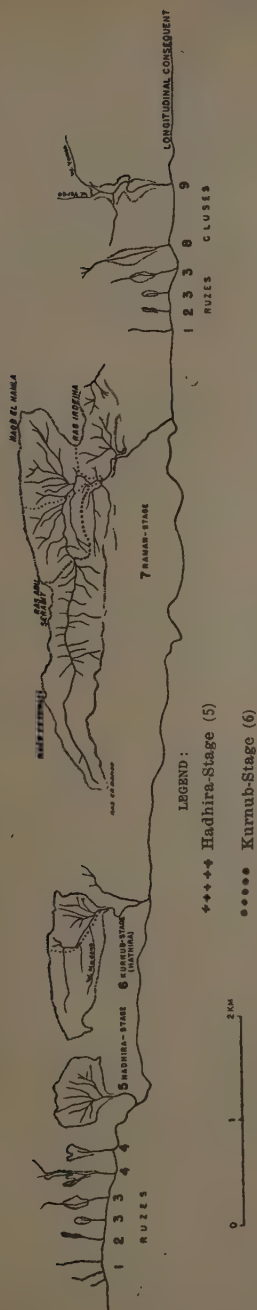
For a clearer understanding of these phenomena we will give a short account of the palaeogeographic development. As demonstrated above, the region of the foothills and the greater part of the western upland up to the Rakhme region were covered by the sea during the Miocene (fig. 4). Eastward, i.e. in the adjoining part of the eastern Negev, as indicated by the Hosb series, there followed a coastal landscape rich in dunes and pools. This coastal region which resembled to some extent the recent North Sinai — El Arish desert, was transversed by rivers descending from the more mountainous Transjordan oldland. With the Pontian upheaval this coastal desert of the Miocene was raised. It became consequently the *Oldland* of the Pliocene coastal plain (foothill-region). Judging from the lateral passage of the Pliocene litoral beds into continental red beds of Kurnub — Hosb type at the Wadi Sab' — Wadi Milh region we may infer a possible low relief of the Pliocene Oldland. This rather arid Pliocene landscape with its low and medium relief was greatly transformed at the end of the Pliocene by a "ground-wave movement". It raised or lowered the folded Negev into regions of up- or downwarps, of anticlinoria and synclinoria, inducing the sometimes pronounced eastward asymmetric turning of the proto-Miocene anticlinal and synclinal elements. The upwarp movement must have followed on very "quickly": for it is noticeable that today the eastern Negev is crossed by neither antecedent nor superimposed riverbeds. The hereditary features of the Pliocene transverse rivers are only preserved east of the watershed: in the foothills and in a part of the eastern uplands. Wherever fold-transversing wadis occur, they join the longitudinal consequents of the strike-valleys as consequents of the youngest cycle.

The low-lying position of the general base-level — the Wadi Araba — gave rise to a more pronounced erosional action than that westwards of the watershed. The anticlines were very soon denuded of the Tertiary cover, leaving only the mantle of capping chert of the Maestrichtian. Eocene, i.e. principally Dano-Montian, remained only

on the synclines; here the Neogene continental Hosb-Kurnub series is still well preserved. The soft nature of these synclinal beds gave rise to vast badland landscapes (see photo II). Capping top-layers of hard middle Eocene are only rarely preserved, and produce that remarkable landmark, the "Madra". The asymmetric and therefore short eastern flanks of the saddles reveal a stepped and denticulated relief in the form of hogbacks and flatirons of hard Turonian limestone and Maestrichtian chert. The asymmetry, and the position of the *local* erosion base, which always lies at the eastern foot of the anticline, gave rise to a much swifter and deeper entrenching of the ravines on the eastern flanks. The short lateral-consequents appear in the form of genuine ruzes or even in the morphologically advanced stage of cluses and finally in small as well as gigantic erosion cirques and funnels. All stages of development from the notch-shaped ruze to the circular Hadhira (radial wadis) to the elliptic Hathira (subsequent-obsequent wadis) and finally to the 33-kilometre long gigantic basin of the Raman can be observed in the Negev. The arid landscape — much better than the humid Jura or the Appalachians — shows up the regular fold-cycle and fold-physiognomy. This accounts for the conception pictured in fig. 6 of the evolution of the most striking relief forms: the simple or normal erosion cirque (Hadhira), the composite erosion ellipse (Hathira), the complex Raman funnel and the cluse (Yemen-Yerqa). (The Raman basin is described below in the survey of the central Negev).

The Erosion-cirque of the simple or Hadhira stage.

Out of the ravines (stage 1 fig. 6) and initial rock-gullies (stage 2 fig. 6) of the asymmetric east-flank, there develop simple ruzes (stage 3 fig. 6) and forked ruzes (stage 4 fig. 6). Lateral erosion induces bottle-neck shaped notches and later the genuine cirque or Hadhira stadium (stage 5). The prototype is found in el-Hadhira of the Jebel el Hadhira anticline ($7\frac{1}{2}$ kms. long, $4\frac{1}{2}$ kms. wide). This cirque has a very elaborate system of dendritic drainage arteries which have penetrated deep into the Nubian sandstone and reflect the bowl-like morphology of this erosion crater. Like the veins of a leaf, the drainage arteries join up in a narrow gorge — comparable to the stalk of the leaf — which finds its exit in the synclinal wadi in the form of a dry water gap. This gorge — a previous ruze — is the only drainage-channel of the Hadhira. Although a number of parallel ruzes have attacked the east flanks of the anticline in the immediate neighbourhood of the cirque they have remained independent ravines, touching the cirque at best at the crêts. They never cut backwards deeply into the bottom of the cirque, and thus played no part in the erosional evolution of the Hadhira. The cirque is by no means the result of the union of several ruzes. The circular shape of the Hadhira with its radial semi-centrifugal pattern points to a structural dependence on a dome-like feature. The Hadhira in fact developed in what was structurally the *highest* and the most arched section of the anticline. It is therefore no coincidence



that the most pronounced upturned hogbacks — up to 60 degrees — and flatirons again occur on that part of the eastern flank of the anticline which belongs to the Hadhira.

The erosion-ellipse of the composite or Kurnub (Hathira) stage.

The erosion cirque of Kurnub, (referred to as el Hathira on the maps (stage 6, fig. 6) is 14 km long and 5 km wide and thus would be more accurately termed an ellipse. The north-eastern third displays the typical branching pattern of the Jebel el Hadhira cirque (stage 5). From this main network a few wadis extend to the south-west end of the erosion-ellipse, running parallel to the strike. These are longitudinal subsequents which, like Wadi Madsus, drain the remaining two thirds of the erosional basin and produce with the wadis of the first third an asymmetric fan-like drainage pattern. Inside the basin, the low monoclinical ridges of the Jurassic limestone and hard quartzitic sandstones of the Lower Cretaceous are the morphologic expression of the subsequent arteries. Neither the ruzes of the east flank (inclining as much as 90 degrees) nor the wadi of the gently dipping western flank (only a few degrees) penetrate to the

Fig. 6. — The Evolution of Erosion cirques.

bottom of the basin. Here again the whole basin drainage has its outlet in a single gorge flowing into the Wadi Yerqa. There we have the most arched and the most elevated raised axis of the Kurnub anticline. As at Jebel el Hadhira, the heaviest assault of headwater erosion was in the structural summit, in a particularly exposed buckle of the Kurnub anticline*.

At first its effect was to hollow out a roughly circular Hadhira; later it extended on one side towards the south-west in the direction of the pitching axis with formation of longitudinal subsequents. The Kurnub stage (6) is, therefore, more advanced in the evolution of the cirques, but still does not attain the "late-maturity" of the more complex Raman stadium to be discussed below.

The anticlinal ridges continuing from Kurnub north-east-ward to Zuweira are also furnished with characteristic ruzes on the asymmetrical eastern flank, the Magrun anticline belongs to this, although it lies east of the watershed, and is part of the Rakhme-anticlinal system. The distinctive morphologic types of the eastern part of the Northern Negev are therefore ruzes and cirques as well as, less frequently, cluses.

The cluse and other transversal breaching valleys.

Genuine cluses are limited to the anticlinal system of Jebel el Hadhira. The simplest form is the almond-shaped indented notch (stage 8, fig. 6) of Wadi Marqab (Wadi Thana of the Newcomb map) north-east of the el-Hadhira cirque. This erosion lens has a double gap at the entrance and exit of the wadi.

Far more advanced in its erosional development is the Wadi Yemen-Yerqa, which is formed by two cluses**.

It begins on the east flank of the Jebel el Hadhira anticline with an almond-shaped incision 200 m deep, formed by one ruze. Like the neighbouring Hadhira cirque, the incision extends down to the Nubian sandstone. The cluse, however, is not closed in the west, but opens out into a second cluse. This second cluse coincides with a small anticlinal bend (west of Wadi Adlikhan) parallel and to the west of the Hadhira anticlinal ridge. Thus the double fold induces the double cluse and the compound bottle-neck valley of the Yemen-Yerqa (stage 9, fig. 6). The longitudinal consequents, the "vals" of Adlikhan and Yemen, lower and upper Yerqa, enter this transverse valley at right angles. (As a result of more recent erosion the main stream of the Yemen-Yerqa has united with the narrow gorge outlet of the Hathira of Kurnub, forming pot-holes and dry waterfalls). The rectangular trellis-pattern is very pronounced in the Yerqa-Yemen drainage system.

* The similarity of the names *Hadhira* and *Hathira* has induced us to use the name Kurnub-anticline instead of the often used Hathira-anticline.

** Marked Wadi Yemen on the Newcomb map; denoted as Wadi Yerqa on more recent maps.

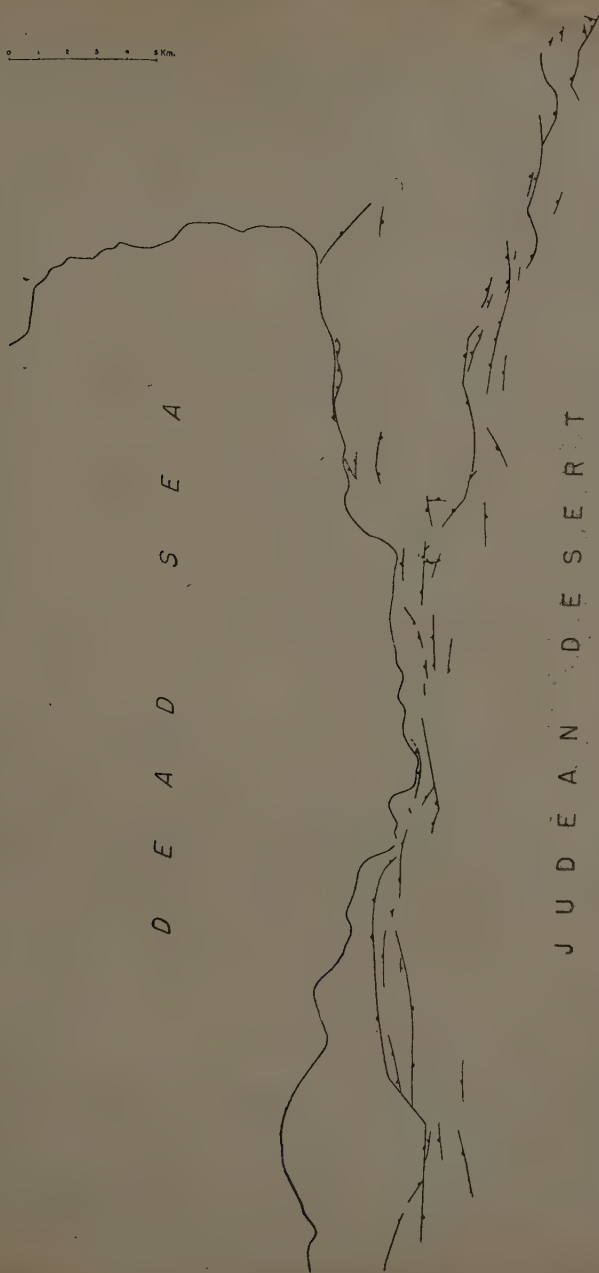


Fig. 10. — Fault-pattern of the flank of the S.W. Dead Sea graben. (surveyed by J. Vroman on behalf of the Jordan Exploration Co. and published by courtesy of Mr. M. Novomeisky).

The *Sanam* plateau (east of the Kurnub Police Post) with its centripetal headwater drainage clearly indicates the basin nature and distribution of the continental Hosb series. The other transverse valleys, such as the gorge of the lower Wadi Murra, and, in particular, the Wadis Amaz, Sutub and Muhawwat, ending in the Dead Sea Graben, are of the parallel-walled canyon type like those of the Judean desert. They demonstrate the rejuvenating influence of Graben-tectonics with waterfall steps, tectonic hanging valleys, etc. The union of the longitudinal "vals" of the Wadi Juraba and Wadi Fai with the upper course of the Wadi Muhawwat still produce a fold-connected rectangular drainage pattern.

Fault-Conditioned Morphology (fig. 10)

Fault-block morphology is clearly noticeable in the Negev at the very edge of the Dead-Sea Graben or of the southern Araba Graben in the form of steplike blocks framed by curved faults. In the fold-Negev proper, on the other hand, faults do not play an important rôle in the relief. With the exception of the horizontal displacements in the transversal valley at Kurnub, it is merely a matter of small transversal dip slips of slight throw crossing the anticlines. The filling up of the transversal depression at Kurnub by Neogene Hosb sands infers a Miocene origin of the horizontal displacements. Most of these faults are typical adjustment-faults of the early Miocene folding and are, as such, confined almost entirely to the anticlines. The strange, meandering course of the fault-line of the Beersheba basin on recent maps (Shaw & Glick) is very improbable as a fault pattern and is in fact not observed in the field. Although the writer assumed a Miocene Graben origin for the Beersheba basin (1931)*, in the light of our present-day knowledge the basin can be better explained by dipping down on all sides of the Hebron and Kurnub upwarps and the synclinal structure thus involved. Pleistocene faults being rare in the mountainous Negev, one can scarcely expect young fault morphology. In its place, fold-structure and fold-morphology stand in closest relation to each other as cause and effect.

II. THE CENTRAL NEGEV UPLAND

a. *General*

From the morpho-tectonic point of view the northern Negev is best divided from the central Negev by the line of Wadi Murra — Wadi Merzebe. This is also the old pass-road of Derb es Sultane leading down from the Naqb el Ghareb watershed over Ras Merzebe into the Araba. The northern Negev with its north-west — south-east folds

* PICARD, L., 1932, Tektonische Entwicklungsphasen im noerdl. Palästina. Zeitschr. d. Geol. Ges.

of Jebel Beqqar, Rakhme — Maqrun, Kurnub — Zuweira — Hadhira, displays a relatively simple fold-structure and fold-morphology despite its pronounced south-east asymmetry. Although in the central Negev the relief can generally be brought into harmony with the structure, the mutual relationship is more complicated.

The first detail of interest is the pronounced *east-west trend of the folds* in the smaller anticlines: Jebel Tewal el Ein, Humra el Hufir, Tewal en Nefkh and Umm Said.

The Raman and the Ureif have anticlines in their north-east sections which maintain the old north-east trend and which plunge in the same direction towards the Wadi Jaib depression. In the south-west on the other hand, at the approach to the Egyptian border, they gradually turn to an east-west trend of the anticlinal mountain ranges of Aneiia, Burja, Araif en Naja, Hoseira and Kherim; at Jebel Sherif the turn is actually to the north-west (see geolog. map of Egypt 1:500,000). The anticlines of the Sinai coast are the first to show the "Judean" north-east trend on the line Yel-leq — Helal and Meghara — Huleiqat.

The central Negev upland, therefore, appears as an *assembling* region of very high anticlines with a strong east-west trend grouped round the zig-zag Egyptian—Israel border with mountains sometimes attaining heights of over 1000 m. As a result the synclinal parts of the folds here also appear at greater altitudes. Similar to the Raman West-slope, the morphology of these synclinal regions east of the watershed are characterized by plateau escarpments and table mountains of varying size. Senonian-Eocene mesas and buttes distinguish the Jebel Lussan, Meghara, Semawe, Ideid, Jebel Qireiq, Abu Taqiye and the region of Wadi Umm Salih. The structural plateaux, entirely lacking in the northern Negev, are developed on a giant scale in the *cuestas* of the et-Tih plateau in the Sinai. From the Sinai they extend beyond the frontier to the Raman-slope or Nefkh-Abda plateau (see section, fig. 7, 8). Hence the plateau morphology of the synclines frequently alternates with the fold morphology of the anticlines. The physiognomy of the central Negev thus takes an intermediate position between the *cuesta* landscape of Central Sinai and the *Jura* landscape of the northern Negev.

The most important ranges in the central Negev — Raman, Araif en Naje, Ureif — are also characterized by a definite south-east asymmetry. There we find erosion-cirques but rarely *cluses* or *ruzes*. The smaller anticlines at Abu Taqiye, Biyar Ideid and north of Jebel Semawe are mostly symmetrical in structure. Like the other small but asymmetrical anticlines south of Ideid and at Wadi Amud, they do not display a pronounced relief. A distinctive characteristic of the main folds of the central Negev are the longitudinal *strike faults* of the south-east flank or, more rarely, in the crest region of the anticlines. These intensify the east-north-east or east-west trend of the structure. In view of the strong throw at Raman up to 1200 m and the dip of the fault of more than 45 degrees, one would expect upthrust-movements. These might have

originated during the Miocene folding but have been rejuvenated during the late-Pliocene upwarp. The close connection between faulting and folding is also observed in the pivotal faults of the Ureif and Meliha anticlines. The rotation turn in both cases occurred at the transition of the asymmetrical anticlines into a dome-shaped structure. As they, furthermore, do not harmonize either in character or in trend with the step and crescent-shaped faults of the Pleistocene Graben border, the assumption of a Neogene age for the longitudinal faults may be justified.

A further characteristic of the central upland is the frequent appearance of *igneous rocks*. As regards the basalt intrusions in the lower Cretaceous part of the Nubian sandstone of Raman, their sill nature is proved by the contact metamorphosis of both the overlying and the underlying layers. More noticeable is the appearance of felsitic and "porphyric" rocks in the central part of the Raman anticline and in the centre of the Ureif arch. Did these come into play in the recent upthrust and upwarp movement, or did they influence the arching as a rigid *Precambrian* nucleus? In either case, the upwarp movement of Raman seems to have proceeded not without the active or passive participation of the magma. Morphologically, the felsitic Jebel-er-Rishe rises like a *massif* out of the sedimentary mantle of the Raman.

An excursion from the foothills of Jebel Qarn over the Beqqar anticline (which belongs to the northern upland), Nefkh-plateau, Raman-Ureif to the Wadi Jerafi Ubara bend, as illustrated in the sections (fig. 7, 8), below, touches the following morphological regions :—

b. *The Nefkh-Abda plateau*

The plateau escarpments forming the biggest portion of the west flank of the Raman upwarp, has been analysed in our survey of the watershed regions (section 3c) with respect to its morphologic significance. On approaching the watershed of the western rim of the Raman the plateau benches take on a steeper dip and change into cuesta escarpments. The most important cuestas are produced by the hard limestone beds of the Lutetian and Turonian. The cuestas are here responsible for the typical homoclinal valleys and their longitudinal-consequent wadis Ajrem, Nefkh and Hewa. On the downward slope of the plateau they join in a right angle the dendritic drainage network of the Hafir system. The small anticlines of Humra el Hafir, Tewal en Nefkh, Umm Said, with their incomplete radial arteries, make but a slight impression on the general Hafir pattern.

c. *Raman upwarp and erosion funnel (Photo III)*

This gigantic anticline with its pitching ends touching the Sinai border at one end and the Jaib-Fiqra depression at the other, is some 60 km in length. Two thirds of it is occupied by a 7 km wide erosion crater, exposing Mesozoic beds down to the Triassic.

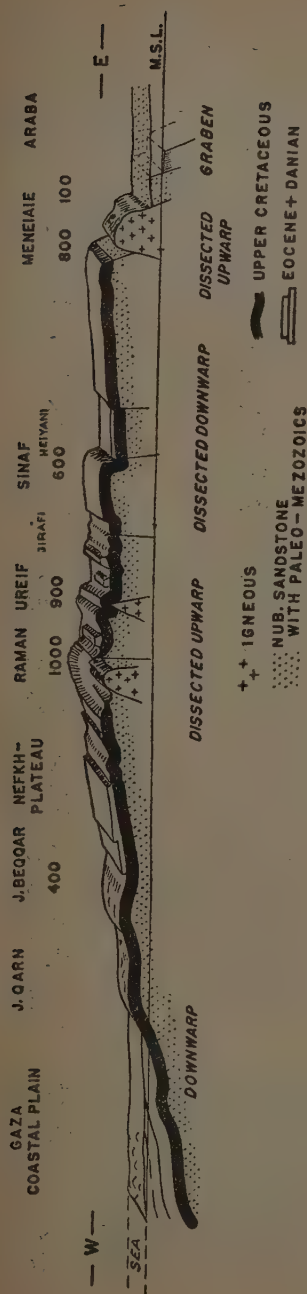


Fig. 7 — Block diagram section through Central and Southern Negev. —

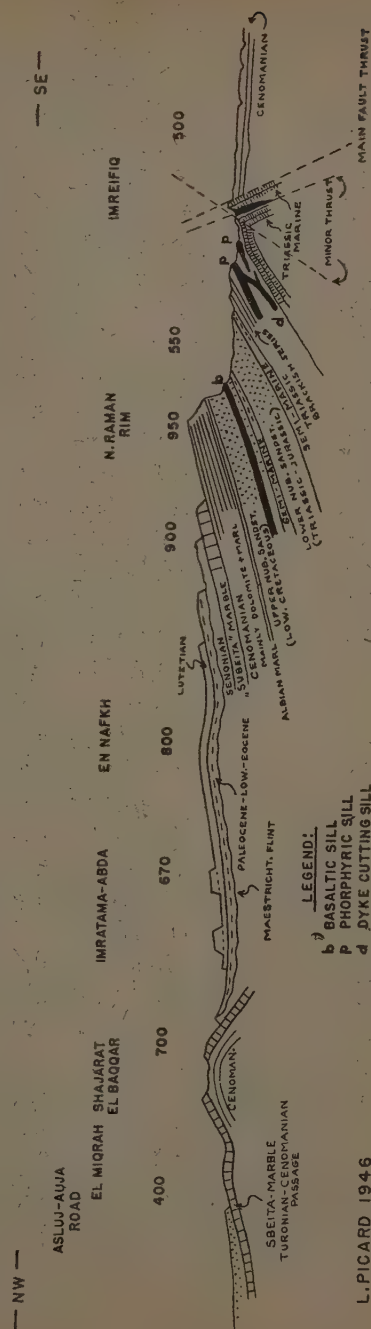


Fig. 8. — Cross-section from the Migrah — Baqqar anticlines through Abda — Nafkh Plateau to Raman.

The dimensions alone indicate that we are not here dealing with a simple erosion cirque or even a composite ellipse. As in the case of these cirques the many-staged erosional evolution of the Raman funnel can again be detected from the drainage pattern (stage 7, fig. 6). The dendritic network of the *north-eastern* half of the Wadi Raman, between Abu es Serabit and Nagb el Hamla — Ras Irdeiha, with the few longitudinal subsequents, is an exact replica in form and pattern of the composite Kurnub erosion fan. Like the latter it has only one exit gorge — the Siq. The north-eastern third of the Wadi Raman (corresponding to the cirque of the Hadhira stage) coincides with the axis of the main anticline (the structural culmination is near the felsitic core). A branching anticline which runs from Imrefig to Ein Jaiadat in the direction of Ras Merzebe, which is carved out by a lesser erosion cirque, is situated between Ein Jaiadat and Ras Irdeiha, and also uses the Siq as a common drainage-outlet. The north-eastern section of the Wadi Raman therefore consists of a *double cirque* corresponding structurally to the above branched anticlines.

The *south-western half* of the Raman funnel from Ras Serabit as far as Ras Raman is much narrower and more elongated in shape. In place of the dendritic radial drainage and the fan shape of the north-eastern half, the south-east section forms a *longitudinal anticlinal valley* drained by a *major subsequent*. Its short north-west tributaries are genuine *obsequents* and its south-west tributaries are *resequents*. The evolution of the Wadi Raman is thus marked by three stages :—

In the first stage a juvenile cirque was formed in the north-east third; it had eroded in all directions and represents the Hadhira (5) stage. Subsequent erosion in one direction towards the south-west led to the elliptic shape and mature stage of the Kurnub-Hadhira (6). In both stages the first half of the Wadi Raman, as far as Ras-Abu-Serabit, was sculptured in this way. Subsequent erosive extension of the south-west half account for the late mature stage (7) of the recent Wadi Raman. The attachment of the south-eastern Ein Jaiadat cirque may have taken place in this last stage. It thus transformed the Wadi Raman from a composite to a complex erosion funnel. Notwithstanding the late-mature stage of the Raman funnel no inversion relief has yet developed as in the case of many Jura or Appalachian mountains. This may be due to the strongly asymmetric structure of all our major anticlines.

d. *Ureif Region*

The complex character of the Wadi Raman morphology is intensified by the longitudinal faults and fissures on the flanks and crests. These form in the eastern flank of the Raman — and particularly on the neighbouring Ureif anticline, a *fault-line escarpment* to which we attribute the lack of cluses and ruzes and the rare appearance of flatirons. Retrogressive escarpment erosion appears to have disposed of the openings of the bottle necks of the earlier gaps and ruze-like notches. The wadis of

the southern flanks fall into the plateaux (synclinal in structure) extending in front of them in the form of open gorges and canyons. Where "dry" waterfalls coincide with the fault-line escarpment, as in the exit of Wadi Ideid from the Ureif ridge, the wadi is morphologically reminiscent of the mouth of the western rivers entering the Dead Sea Graben. This impression is reinforced by the fact that the canyons of the Ureif are also entirely cut in dolomitic Cenomanian rocks.

The Cenomanian canyons then dissect the *table landscape between Raman and Ureif* into the rough "very broken rocky ground" region as indicated on the Newcomb map (1 : 250,000). On its synclinal axis at Imrefig isolated horizontal Senonian table mountains remained above the Turonian-Cenomanian upland plain. Larger plateaux with splendid Eocene escarpments occur in the south-west between Ras-Raman and Jebel Ureif (see fig. 9). These form the Mesas of Jebel Lussan and Jebel Meghara.

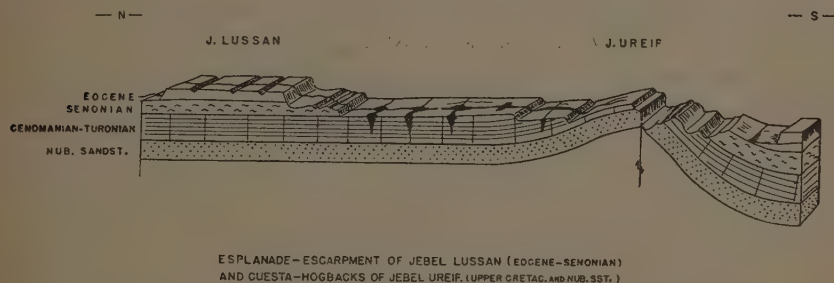


Fig. 9. — Plateau and fold morphology of Jebel Lussan — Jebel Ureif.

The drainage sluice of Wadi Raman, i. e. the gorge of the Siqq, up to 100 m. wide and 400 m. high, also belongs to the plateau between Ureif and Raman. Into the transversal canyon of the Siqq flow longitudinal canyons which are different from the soft "vals" of the northern Negev owing to their uniform gorge-like shape. It is again the outcrop of hard Cenomanian formations which conditions the coarse-rectangular pattern of the Ureif "hinterland", and thus contrasts with the finely etched trellis pattern of the softer rocks of the Dano-Eocene synclinal areas. Jointing and some faulting may have played a role in the development of this rectangular drainage. However, in the development of erosion cirques, like in that of ruzes and cluses, faults have generally had an obstructing effect. Despite the considerable altitude of Jebel Ureif the arch, faulted in its centre, displays only a small cirque. The Nubian sandstone outcrop in Wadi Hadab may have lost its cirque or cluse character on account of the fault-line escarpment morphology.

c. *Jerafi downwarp*

Descending from the central anticlinal regions of the Raman and Ureif into the southern Negev, the heights drop rapidly from an average of 800 m down to 300 m at the bend of Wadi Jerafi (Bir Ibn Auda). We again find ourselves — but this time east of the watershed — in a region of synclinal structure of regional magnitude. In place of the rectangular pattern of Raman — Ureif we again meet with a coarse-textured dendritic network of wadis — the Jerafi with its main branches Umm Athla, Ubara, Abu Taqiya.

Just as the fold-ranges of the upwarp region of the central Negev link together at the watershed of Jebel Lussan, so inverse folds turn down in the opposite direction from all sides to the downwarp centre at Ubara and to the Jerafi bend. The folds of this regional basin are mostly small, shallow and *symmetrical*, similar in type to those met with in the other structural depressions (Khalasa — foothills, Jaib — Fiqra depression). Small longitudinal subsequents lead to the strike valleys of Umm Sheisha, Upper Wadi Amud and Wadi Isheishiya, thereby to a certain extent upsetting the general coarse-textured dendritic pattern of drainage. The little folds do not always appear on the geological survey maps nor on the topographical maps, but are distinguishable on aerial photographs at Jebel Ideid, Jebel Abu Taqiya and Jebel Qureiq.

In the central part of the downwarp is the undrained playa, the Qa'at el Quireiq of pre-Pleistocene age (intersected and gravelled by consequent diluvial wadis), and there are Neogene basin sediments (Hosb series?) at Ubara, and gently dipping Paleocene Taqiya marls. The Eocene extends as a tabulary landscape between Jebel Ideid and Jebel Taqiya (see general cross section). The region between Ideid and Sinaf and round the Qa'a of Quireiq is a flint-strewn Hamada (with Maestrichtian-Campanian subsoil) on which relics of Eocene stand out in relief. These are the mesas and buttes of Wadi Amud, Wadi Irbab, Jebel el Quireiq. The dislocation of Ajramiya — Ubara — lower Wadi Jerafi, comparable to a rotation fault, has had no significant influence on the consequent drainage pattern. The lower Wadi Jerafi belongs to the Meliha — Sinaf region, a region best assigned to the southern Negev.

Summing up the main morphotectonic features of the central Negev, we distinguish an orographically, as well as structurally, elevated middle section — the Raman-Ureif upwarp with a distinct fold relief and fold pattern encircled in the east and the west by downwarps. The morphologic character of the downwarps is shown in structural plateaux and escarpments with small and usually symmetric folds of low relief. Both synclinal regions are distinguished by a roughly dendritic wadi pattern: the Auja-Hafir system in the west, the Jerafi system in the east.

The eastern synclorium unites with the Fiqra-Jaib depression to form the large Wadi-Jaib — Wadi Araba depression which has hitherto displayed no Graben tectonics at the surface. The other faults of the central Negev — particularly the Ra-

man-Ureif culmination have exerted but insignificant influence on the main morphologic features despite their fault-line escarpments.

III. THE SEDIMENTARY SOUTHERN NEGEV

From the headwaters of Wadi Heiyani there starts an anticlinal range of *Jebel es Sinaf* tending first north and then north-eastward, which afterwards bends round into the east-west extending anticline of *Jebel Wadi Hamth*. In 1943 we assigned the latter anticline (our former Baha anticline or Maliha anticline of the P.P.D., see Shaw 1948) to the Jerafi synclinorium. The Sinaf does disclose, however, a typical northern or north-north-east trend more common for the southern Negev ranges of Igfi and of the rim of the Araba. The Sinaf anticlinal presents a range with altitudes up to 600 m, of considerable length and with a pronounced asymmetry towards the east amounting to almost 90 degrees. It can therefore scarcely be ranged with the relatively small and low asymmetric folds which distinguish the Jerafi downwarp. The Sinaf must be regarded either as an independent fold range or must be attached to a new upwarp system. But this new upwarp system attains its maximum height of 800 m in the *Jebel Abarikhat* south of Wadi Igfi. There it reveals an erosion cirque of which only the western half is preserved as a result of the down-faulting of the Araba Graben. This *half-cirque* is drained by the various wadis of *Meneiaie* and exposes a pre-Cambrian crystalline nucleus with a mantle of Paleo-Meozoic Nubian sandstone and intercalations of marine Carboniferous*.

If we include the Sinaf-Hamth anticline in the Meneiaie upwarp, the Sinaf would assume a similar structural position as for instance the *Jebel el Hadhira* anticline or the *Rekhme* anticline with regard to the Kurnub upwarp. In other words, the Sinaf would belong to another *anticlinorium*, the anticlinorium of the southern Negev culminating around the heights of Meneiaie. The great depression between the Sinaf and the Meneiaie heights, however, far exceeds in width the normal Negev synclines, e.g. the synclines of Wadi *Rekhme*, *Sanam* and Wadi *Fai*. It assumes almost synclinorial dimensions. Characteristically enough this *Heiyani-Igfi* "downwarp" does not reveal the trellis pattern of ordinary synclines but the coarse-textured *dendritic* wadi pattern of genuine synclinorial regions. The wadis of Heiyani-Igfi again drain extensive *plateaux* whose Senonian subsoil is covered with fragments of Maestrichtian flints, the

* The Carboniferous of the manganiferous limestone of Wadi Meneiaie (see discussion in Shaw p. 29) is demonstrated by our find of a *Productus* (in 1949), thus comparable to the Naab limestone of the Sinai. The hypothesis of Wellings-Willis of a gigantic horizontal displacement of Wadi Araba based on the assumed marine Cambrian in the Wadi Meneiaie therefore loses one of its main arguments.

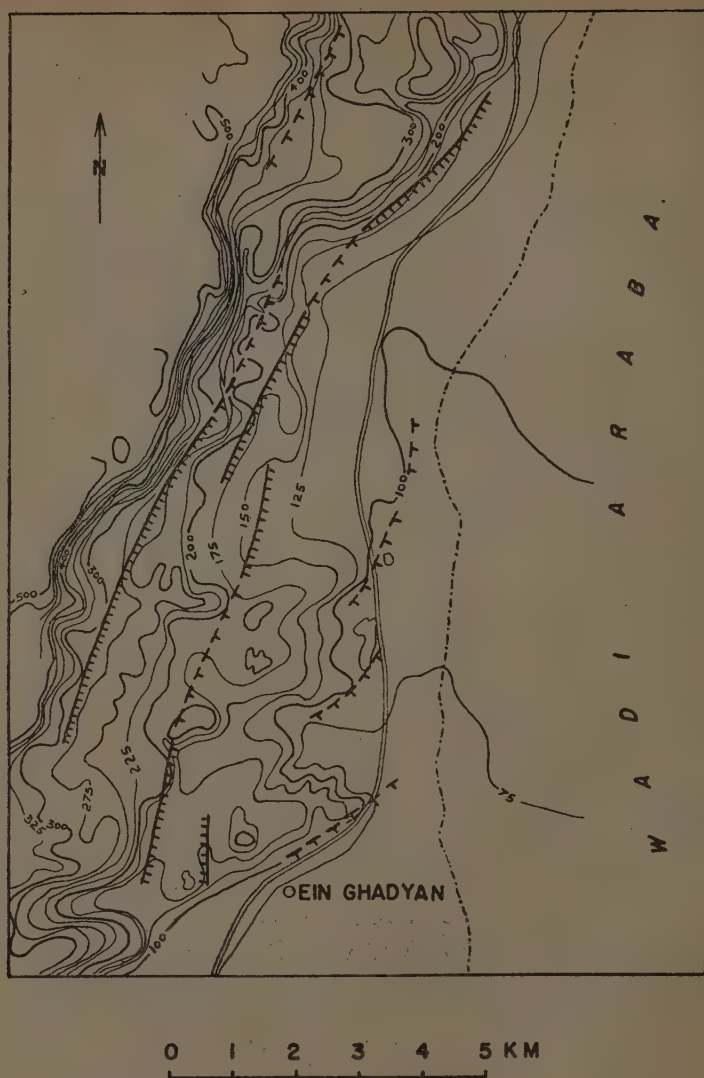


Fig. 11. — Part of fault pattern on the Western flank of the Southern Araba graben.
(surveyed by S. Shifan on behalf of the Geology Survey of Israel).

"broken flat gravel" of Newcomb's map. Only occasional relics or outliers of Eocene have remained on these Hamadas. Structurally, topographically and hydrographically both the Heiyani and the Jerafi downwarp unite at the Ein el Meliha spur and pass from there into the giant Araba depression of the Wadi Jaib. Although the union of the eastern and western Wadi Sehir temporarily produces a rectangular course in these wadis, the principal drainage pattern of Sinaf and still more that of its continuation as far as Jebel Abu Hamth (Meliha) is more of the radial type. Another remarkable drainage feature at Jebel Sinaf is the appearance of a genuine *anular subsequent valley* behind the flat-irons of the eastern ridge. The peculiar character of the Sinaf as to both morphology and hydrography might be explained as a *young domal uplift* at which the older east asymmetry is still preserved.

The fault of the lower Wadi Jerafi is presumably connected with the asymmetric northern flank of the Wadi Abu Hamth anticline, distinguished by hogback ridges and small longitudinal subsequents. The fault rotates at a structural high-point (exposing here Nubian sandstone). The rest of the anticline becomes more and more symmetrical and finally dips at Meliha, near the 200 m contour below the Alluvial of the Wadi Araba. No visible Graben-faulting is noticed here. The various small faults which transverse the Meliha portion of the anticline are of the adjustment type connected with the old Miocene folding.

The wadis of the western heights and slopes of the Meneiaie upwarp are drained by lateral consequents, which form typical Cenomanian canyons and terminate in the undrained alluvial plain of Igfi — morpho-tectonically allied to the Qaat-el-Quireiq. The influence of Graben tectonics is only noticeable at the approach of the crêts overlooking the Araba and the 400 m high Cenomanian cliffs. Raised and sunken blocks, small Graben and horsts, step — and crescentic faults have so forcibly dissected the border region of the Graben, (as may already be observed on the old Hull map) that parts of the eastern Meneiaie upwarp, are now below the Araba plain.

In the extreme arid climate of this southernmost Araba valley appear large alluvial fans, giant playas and inland dunes. Wind increasingly becomes the dominating atmospheric agent. Fault tectonics prevail over fold tectonics, fault-block morphology predominates over fold morphology, crystalline rocks gradually replace the sedimentary rocks (Picard 1941). The southern *sedimentary* Negev changes into the southern *crystalline* Negev, the northernmost outpost of the crystalline Sinai (Photo I).

REMARKS ON LITERATURE

Very little has been published on the structure and still less on the morphology of the Negev. G.S. Blake in his earlier (1928) and more recent (1947) "Geology and Water Resources of Palestine" (Jerusalem, Government Press) speaks of the "monoclinical" steps and folds of Kurnub and Raman. D.H. Kallner in "Outlines of the Geomorphology of Judea" (Bull. Soc. Roy. Geogr. d'Egypte Vol. 21 1943 p. 35) follows this concept. In our "Structure and Evolution of Palestine" (Bull. Geol. Dep. Hebrew University, Vol. 4. Jerusalem 1943 No. 2-3-4) we discussed and differentiated the main structural elements: asymmetric anticlines, up — and downwarps, and fault-blocks. In "The Precambrian of the North Arabian — Nubian Massif" (Bull. Geolog. Dept. Hebrew University Jerusalem 1941 Vol. 3 Nos. 3-4) we had previously dealt with the crystalline region near the Gulf of Aqaba. Based on the

geological survey of the Palestine Petroleum Development Co. (a subsidiary of the Iraq Petroleum Co.), on interpretations of aerial photographs by Glick, on his and Mitchell's observation near the Gulf of Aqaba, S.H. Shaw compiled and published the "Geological Map of Southern Palestine" with Explanatory Notes (Jerusalem 1947, Govt. Press). Reference is made to B. Willis' "Wellings observations of the Dead Sea Structure" (Bull. Geol. Soc. of America Vol. 49 pp. 659).

A valuable aid in the detection of fold drainage patterns, ruzes, cluses, cirques are the so-called Newcomb maps published by the British General Staff Office, London, 1915, the Northern Negev sheets of the 1:100,000 edition of the Mandatory Government of Palestine, and the partly new and partly revised 1:100,000 maps of the whole Negev prepared by the Survey of Israel.

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LONG RANGE FORECASTS OF RAINFALL

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The meridional shift of the anticyclonic cells of the subtropic latitudes during the year suffers deviations assumed to persist for several months; these are believed to be of prognostic value for winter rain in Israel. The influence of the position of the anticyclonic cell in the Middle East is scrutinized, and a significant negative correlation is found between rainfall amounts along a meridional section in the Sudan during August-September and at eight representative stations in Palestine during the following winter.

During the last seven years methods of long range forecasting have been developed by meteorologists who are now members of the Meteorological Service of Israel. Considerations of general circulation in the belt of the Horse Latitudes and its neighbourhood conditioned the choice of the first method to be investigated. The stationary anticyclone covering this area shifts northward during summer in the northern hemisphere and southward during the northern winter. The movements of the anticyclone are not equal in range and time from year to year, and the position of the anticyclone may be too far north or too far south for a considerable time; the reasons for these deviations are not yet fully understood, but the existence of the deviations and their persistence during considerable time-intervals (months, quarters and longer) are well-known facts.

Israel is situated in the northern part of the belt of the Horse Latitudes. The northward movement of the anticyclone in the northern summer brings the whole Levant area within its influence with consequent fine weather. In winter the anticyclone moves southward, and upon the amount of its movement to the south depend the weather characteristics of the winter in Israel. A move of the anticyclone far to the south gives way, in the Mediterranean, to the development of the belt of the Westerlies with its wandering cyclones, bringing ample rainfall to Israel. On the other hand, a position of the anticyclone too far north in winter leaves Israel partly under its influence with consequent scarce rainfall.

If we assume that the displacement of the anticyclones is generally slow and persists uniformly during a greater part of the year, we may expect that a displacement in winter had started some months before, say in August or September. Verification of this may be of value in forecasting the rainfall in Israel during the following winter.

For this purpose, one way of ascertaining the displacement of the anticyclone in late summer was developed by analysing Sudanese rainfall in August and September. Thus, as ample rain during these months is evidence of a northward displacement and and vice versa, an inverse relationship would appear to exist between late summer rainfall in the Sudan and Israel rainfall during the following winter, and an examination of August and September rainfall data at Sudanese stations should be of value in forecasting.

On this basis, data on Sudanese rainfall in August and September for the period 1924-1946 were collected and the correlation was tested during 1942 to 1949 when actual rainfall was compared with the forecast. The main obstacles were the shortness of the series of observations at both ends and the fact that the period of comparison had to be restricted to the years 1924 and onward. In spite of this, a significant negative correlation was found between the sum of rainfall amounts in August-September at four Sudanese stations distributed fairly equally between 18° and 5° N along a line extending south of Israel, and rainfall totals, during the following winters, of eight representative stations distributed equally over the area of Palestine north of Beersheba.

It is one of the characteristics of this correlation that its coefficient is significantly higher when only years with deviations from the mean by more than 20% of the rainfall in the Sudan are considered, i.e. years with a considerable displacement of the anticyclone. Thus correlation coefficient for the whole series 1924-1946* is

$$r = -0.432 \pm 0.114,$$

whereas the correlation coefficient for years with deviation of more than 20% is

$$r = -0.674 \pm 0.137.$$

* Another fact pointing to the inverse relationship between rainfall during August and September in Sudan and that during the following winter in Palestine may be found in comparison of the relative skewness of the dispersion of the two series of data when arranged in ascending order. When considering the whole series available for both countries, the following values for relative skewness are obtained:

$$\text{Sudan: } s = +0.24; \text{ Palestine: } s = +0.16.$$

But when again considering years with deviations from the mean of more than 20 per cent of rainfall only, the relative figures for the skewness change in the following way:

$$\text{Sudan: } s = -0.33; \text{ Palestine: } s = +0.35.$$

This similarity in the amount of skewness of the two series compared, with opposite sign, may be regarded as indicative of an interrelation between them. The author is well aware of the shortcomings of this approach, owing to the small number of cases involved (7 for the Sudan, 11 for Palestine). Relative skewness was computed according to the following formula:

$$s = \frac{m - m_1}{q_1 - q_3}$$

where: s = relative skewness,
 m = mean,
 m_1 = median,
 q_1 = upper quartile,
 q_3 = lower quartile.

Thus the forecasting value of the correlation is much higher in years of large deviations in the Sudan. For normal years, additional correlations must be sought in order to strengthen the forecasting value of the present correlation.

The correlation described above was devised in 1942; publication of the trial forecasts began in 1946. Four out of seven forecasts were fully borne out; the three misses occurred when rainfall in the Sudan was normal.

The following table shows the forecasts and their results :

| YEAR | FORECAST | RAINFALL | QUALITY OF FORECAST |
|--------|------------------|--------------------------|---------------------|
| 1942-3 | <i>normal</i> | <i>much above normal</i> | — |
| 1943-4 | <i>normal</i> | <i>normal</i> | + |
| 1944-5 | <i>normal</i> | <i>much above normal</i> | — |
| 1945-6 | <i>minus 20%</i> | <i>minus 15%</i> | + |
| 1946-7 | <i>minus 35%</i> | <i>minus 35%</i> | + |
| 1947-8 | <i>normal</i> | <i>normal</i> | + |
| 1948-9 | <i>normal</i> | <i>much above normal</i> | — |

In conclusion, it may be assumed that the relation found between deviations in rainfall in the Sudan and inverse deviations several months later in Israel is a proof of the persistent blocking of the general circulation by shifts of anticyclonic cells poleward or equatorward for several months. The fact that correlation was best along a meridional line may hint at the adherence of the blocking cell to a certain longitude, possibly owing to geographical influences. Further investigations, especially with the aid of data on upper air now being compiled in the Middle East, may throw more light on these oscillations and the way in which they may form a bridge between the mechanism of synoptic developments on the one hand and that of secular fluctuations on the other.

This note is published with the permission of the Director, Meteorological Service; most of the data on which it is based were obtained with the kind assistance of Mr. A. W. Ireland, Government Meteorologist, Sudan.

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THE FORMATION OF "SALT TABLES" IN NATURAL AND ARTIFICIAL SOLAR PANS

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The surface of salt deposits in natural or artificial solar evaporation pans is usually very uneven. Small objects such as rods, twigs or stones protruding from the bed are quickly covered with a salt crust which thus enlarges the protrusion. Ripples, similar to those occurring in sand in the shallow waters of seas and lakes, are produced in solar pans, and these too are exaggerated by salt deposits. (Fig. 1)



Fig. 1.a—Salt deposits in solar evaporation Pan

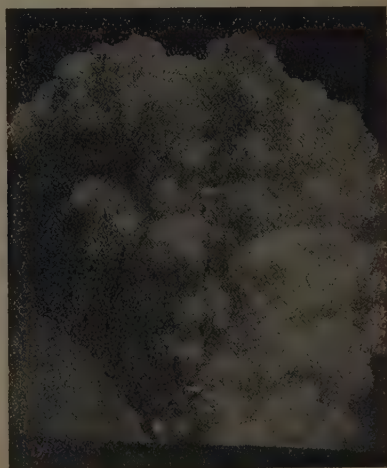


Fig. 1.b — Protrusion enlarged by Salt crust

When a rough-surfaced layer of salt is washed by rains it develops an extraordinarily smooth surface. The salt expanse of Lake Bonneville, Utah, for example, becomes so smooth after the winter rains that it has become the favourite terrain for attempting world speed records. This remarkable smoothness appears also on the salt pans at the

* Died December 1948

Dead Sea when they are washed either by rains or by fresh water which is pumped into the pans to prepare them for a new salt-crop.

An investigation into the mechanism of the dissolution of salt in solar pans, and into the reason for the smoothing of previously rough surfaces, resulted in the following observations.

A layer of water covering a smooth salt surface develops a stable stratification, the salt-concentration being high near the surface of the salt and low near the surface of the water. This stratification is not affected by temperature changes, since the differences in the specific gravity of the layers are sufficient to maintain stability even under extreme conditions. (Temperatures of 60°C due to radiation of the sun have been observed in the bottom layers.) The solid salt dissolves into a stratified layer of this kind only by *diffusion*.

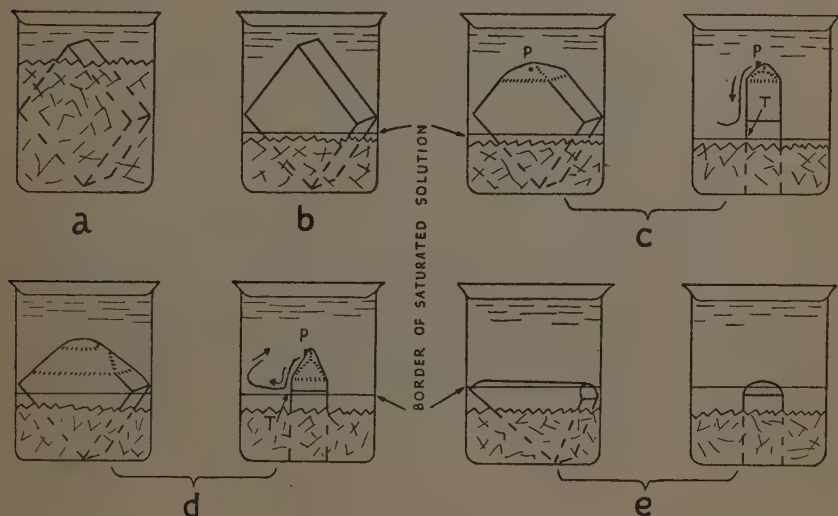


Fig. 2

Convection of the various strata only occurs when the salt surface is rough or when protrusions such as those described above project into the unsaturated strata of the supernatant salt solution. In such cases a film of saturated brine is formed which flows down the sides of the ridges, quickly reducing their height. Dissolution does not occur along the sides of the ridges, which are protected by the descending brine film, but the ridge tops quickly dissolve, flattening to form "tables". When these tables are reduced to the level of the surrounding salt floor, the convection stream is halted and a perfect levelling of the surface, often covering many square miles, results. Further dissolution can take place only by the much slower process of diffusion.

The above phenomenon can be illustrated by a simple experiment: a flat, rectangular salt crystal (about $4 \times 6 \times 1$ cm) is placed in a beaker (see Fig. 2a) and almost completely buried in coarse salt; the beaker is then cautiously filled with water. During the first five minutes, a considerable part of the coarse salt dissolves, forming a saturated layer which, together with the remaining solid salt, surrounds the lower half of the crystal (Fig. 2b). During the next two hours, the crystal gradually dissolves, passing through the stages indicated in Figs. 2c and 2d. After twenty-four hours it is reduced to a flat "table" (Fig. 2e). The course of this process may be clearly observed if a small

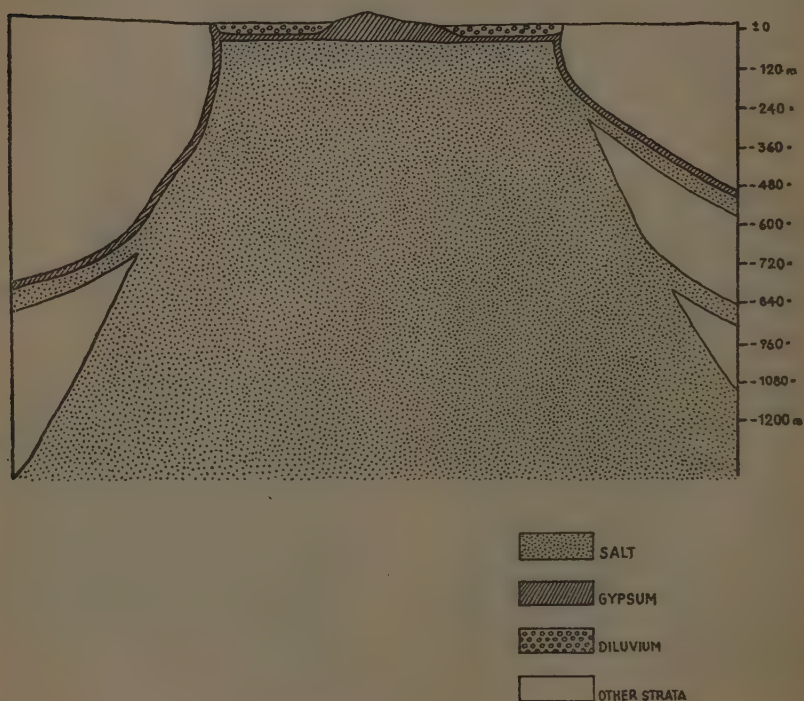


Fig. 3

crystal of potassium permanganate is placed on the top of the salt crystal (*P* in Figs. 2c and 2d) with the aid of a small wire hook. As the permanganate dissolves, a thin red stream flows slowly down the sides of the salt crystal to the point *T*, where it turns

sharply away, flowing slightly upwards. When the salt crystal reaches the stage shown in Fig. 2d, the coloured stream turns again at point *S*, and flows back to point *P*. Thus the path of the salt, as it dissolves from the salt crystal and flows down the slope of the peaks and ridges, may be vividly demonstrated.

These observations may explain the appearance of "salt tables" (Salzspiegel) often found in geological salt formations. Fig. 3 shows such a table found in Lüneburg. A similar formation occurs in Djebel Usdum.

Such "tables" can only have been formed during a period when the salt deposits were covered by a body of water in which convection streams could develop freely under the influence of gravity, but which did not seep appreciably through the salt mass. The covering water layer must have consisted of strata of differing concentration.

These observations may also explain another characteristic form of natural salt deposit, which occurs as a sequence of alternating thin and smooth layers deposited one over the other. It is probable that these layers formed by evaporation were originally rough and were washed to smoothness by standing water accumulated, e.g., during winter rains. This hypothesis is supported by the fact that such smooth salt layers are often separated by a stratum of fine clay, in all likelihood deposited when the standing water was muddy.

THE IODINE NUMBER ESTIMATION OF BITUMEN

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The mechanism of HBr formation during bromination of Bitumen was studied.

It was found that the formation of HBr is due to a secondary reaction: decomposition of the primarily formed addition products.

This reaction is reversible and can be controlled by adding HBr prior to the bromination.

Thus, the Total Bromine (or iodine) numbers as indicated by the amount of absorbed bromine is also the true iodine number which expresses unsaturation of the bitumen. No reduction on account of the formed HBr is required.

Whilst there is a great deal of descriptive literature on the oxygen absorption of bitumen, nothing is known as to the mechanism of this process. The obvious parallel with the oxygen absorption of drying oils makes it probable that in bitumen, too, the oxygen absorption is connected with the presence of unsaturated molecules.

The present investigation is aimed at developing a reliable and simple method for the determination of unsaturation in bitumen. The material used was a commercial product ([Mexphalt] 80/100); its properties are summarized in the Appendix.

In petroleum products the determination of unsaturation is by halogenation. The results are often ambiguous because the reaction is accompanied by evolution of hydrogen halides. In the bromination, for example, HBr can result from four different reactions:

- (a) secondary decomposition of the primary addition product
- (b) substitution
- (c) hydrolysis of the addition or substitution product when it comes in contact with water
- (d) reaction with bromine of sulphur-containing impurities of the material to be tested.

In order to characterise the unsaturation of a product such as bitumen, it is obviously necessary to determine not the "total halogen number", but the true "addition halogen number".

Poell¹, in his critical study of the iodine number of natural and petroleum asphalts, reviewed the various methods in use for the determination of the iodine number of mineral oils and bitumen. He found that the McIlhiney method* gave the most consistent results.

To avoid hydrolysis, he brominated in a tightly closed Claisen distilling flask and then distilled in vacuo the whole of the surplus bromine, HBr, and carbon tetrachloride into a solution of KI. The bromine and the HBr were titrated in the KI solution. Thus the brominated product left in the distilling flask had no contact with water. In all the tests carried out using this method on bitumen and mineral oils, he found that the total iodine number (B) was exactly twice the HBr value (C), and he concluded that only substitution, and no addition, occurred. He furthermore proved that no organic acids (water-insoluble) were formed during the bromination of bitumen.

Since it is also possible that HBr is formed from the brominated product by decomposition during distillation, Poell tested the distillation residue for its iodine number; he found it much lower than the first iodine number (8.8 as compared with 160), and again the HBr was exactly half the total iodine number. Poell therefore claims that, if HBr were formed by decomposition, unsaturated bonds would be regenerated and the new iodine number would be of the same dimension as the first. As this is not the case, he comes to the conclusion that *natural mineral oils and bitumen do not contain aliphatic double bonds*. We shall return to this contention below.

Three different methods of iodine number estimation were tried first: the Kaufmann method, the Hanus method and the modified McIlhiney method.

The Kaufmann method showed a negative value for the true iodine number (C being more than B/2), while the Hanus and McIlhiney methods gave positive values (in contradiction to Poell's findings).

We — like Poell — found the McIlhiney method to be the most convenient and this method was adopted throughout this work.

To avoid loss of HBr evacuated vessels of different form and size were used (first proposed by Crossley-Renouf²).

We may summarize the results as follows:

Influence of Apparatus Used: No appreciable differences in (B) values were found when using different types of apparatus, all other conditions being the same, whilst (C) values vary with different kinds of apparatus.

* In this method CCl_4 is used as solvent for both bitumen and bromine.

1. POELL, H., 1931, Die Jodzahl von Natur- und Erdoel-asphalten. *Petroleum*, **27**, 817—826.
2. CROSSLEY-RENOUF, 1908, *Soc.* **93**, 648.

Reproducibility: Whenever the same conditions were maintained, good reproducibility for (B) values was obtained, whilst the (C) values (and consequently the (B-2C) values) showed poor reproducibility.

Influence of Reaction Time, and Bromine to Bitumen Relation:

(B) increases with time and bromine-to-bitumen relation in a smooth curve. (C), whilst rising, shows a very inconsistent curve and generally reaches a constant value before (B). The increase of (C) is much smaller than that of (B).

Generally speaking, (B) values are reproducible and constant, depending clearly upon reaction time and bromine-to-bitumen relation, while (C) values are much less dependent on these factors and seem to depend also upon others which were not controllable in our tests. (C) values do not seem to follow the same trend as the (B) values.

The answer to the problem was found in the elucidation of the origin of the hydrogen bromide liberated during bromination. It was first shown by comparing the titration and the precipitation figures, that all the acid formed was actually hydrobromic acid.

TABLE I.

HBr in mg

| By Titration | By Precipitation |
|--------------|------------------|
| 29.4 | 31.5 |
| 15.1 | 15.0 |
| 17.8 | 17.5 |
| 15.0 | 14.9 |

A detailed study was then undertaken of the behaviour of (B) and (C) under different conditions.

(a) *Addition of HBr prior to titration*

After completion of the bromination reaction, a measured amount of aqueous HBr solution was run in, well shaken, and KI solution added. Then the content of the flask was titrated for Br₂ and HBr (Table II). The (C) values are the corrected for added HBr.

TABLE II.

Bitumen used: 360 mg, Concentration of Br₂ Solution: 26 mg per ml

| Time in Minutes | Added HBr ml n/10 | (B) | (C) | (B-2C) |
|-----------------|-------------------|------|------|--------|
| 15 | 0 | 77.5 | 30.5 | 16.5 |
| 15 | 10 | 77.0 | 29.2 | 18.6 |
| 15 | 40 | 79.0 | 26.3 | 26.4 |
| 15 | 108 | 75.5 | 21.2 | 33.1 |

No appreciable change in the (B) values occurs; but the (C) values decrease significantly for large quantities of added HBr.

(b) *Elimination of HBr during the bromination reaction by a current of nitrogen*

The apparatus used is given schematically in Fig. 1; its operation is self-explanatory.

(i) Influence of increasing duration of flow of the gas.

TABLE III.

Nitrogen Flushing Experiments

Bitumen : 360 mg Bromine solution : 15 ml

Concentration of Bromine solution : 15 mg Br/ml

| | Time of Reaction Prior to Gas flow in min. | Time of Gas flow in min. | Distillate Residue | | | | (B) | (C) | (B-2C) |
|----|--|--------------------------|--------------------------------|------------------|-------------------|------------------|------|------|--------|
| | | | HBr ₂ ml n/10 | Br ml n/10 | HBr ml n/10 | Br ml n/10 | | | |
| 1. | 10 | 10 | 5.7 | 1.0 | 1.7 | 27.2 | 67.0 | 25.7 | 15.6 |
| 2. | 0 | 20 | 6.9 | 2.4 | 1.6 | 24.5 | 71.5 | 29.2 | 13.1 |
| 3. | 0 | 60 | 8.0 | 2.8 | 1.2 | 23.7 | 73.0 | 31.6 | 9.8 |
| 4. | 0 | 120 | 9.7 | 10.0 | 0.8 | 17.3 | 70.0 | 36.1 | -2.2 |

(ii) In Table IV an experiment is summarised which lasted six hours (at constant rate of flow) : every hour the amount of bromine and of HBr present in the distillate and in the residue, respectively, was titrated.

TABLE IV

Nitrogen Flushing Experiments

Bitumen : 360 mg

Amount of bromine : 15ml, 13 mg/ml

| Time (in min.) | Distillate ml n/10 | | Residue ml n/10 | | (B) | (C) | (B-2C) |
|-------------------|-----------------------|-----------------|--------------------|-----------------|------|------|--------|
| | HBr | Br ₂ | HBr | Br ₂ | | | |
| 60 | 7.9 | 8.2 | | | | | |
| 120 | 2.1 | 5.2 | | | | | |
| 180 | 0.6 | 0.9 | | | | | |
| 240 | 0.85 | 2.7 | | | | | |
| 300 | 0.3 | 2.6 | 0.84 | 5.5 | 87.0 | 43.8 | -0.6 |

(iii) In the experiment characterized in Table V the bromination reaction was allowed to proceed for 30 minutes before nitrogen was passed through the system. This was continued until no more HBr came over.

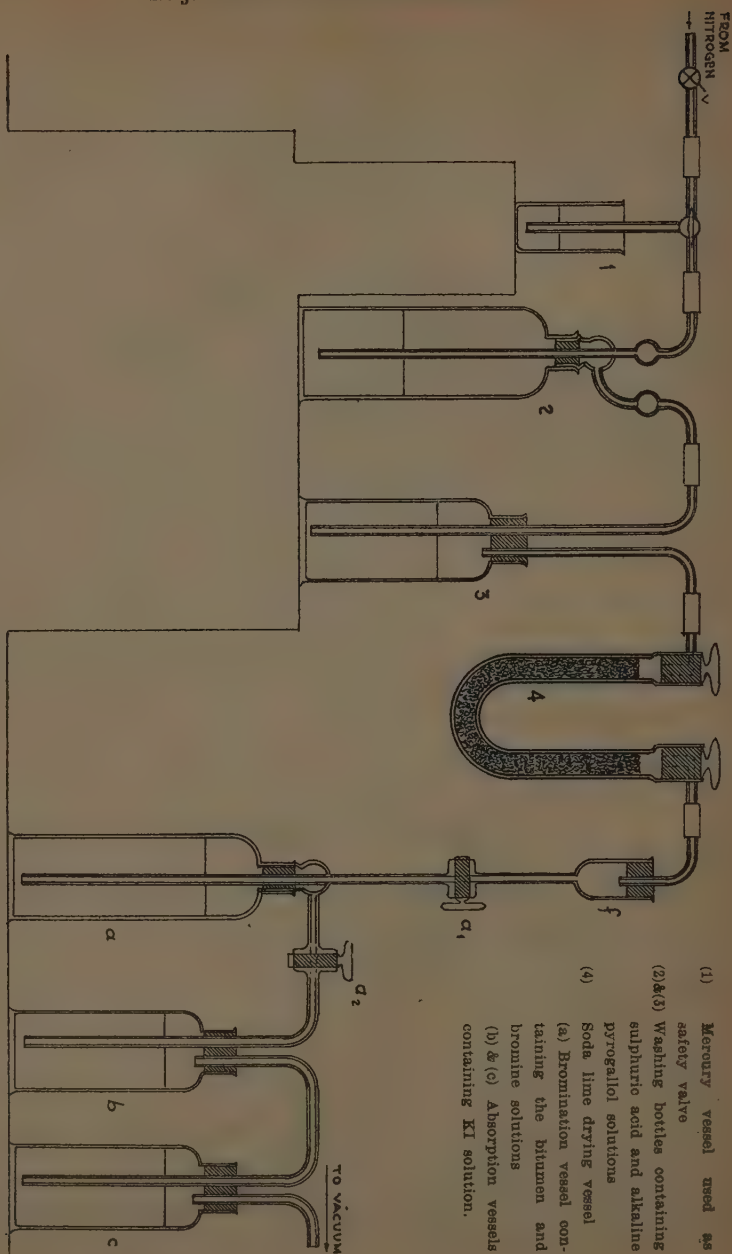


FIG. 1

NITROGEN FLUSHING APPARATUS

TABLE V

Nitrogen Flushing Experiments

Bitumen : 360 mg

Bromine solution : 15 ml, 26 mg/ml

| Time of N ₂ Flow (min.) | Distillate (ml n/10) | | Residue (ml n/10) | | (B) | (C) | (B-2C) |
|--|-------------------------|-----------------|----------------------|-----------------|-----|------|--------|
| | HBr | Br ₂ | HBr | Br ₂ | | | |
| 90 | 12.8 | 24.9 | | | | | |
| 210 | 2.0 | 17.5 | | | | | |
| 270 | 0.5 | 6.8 | | | | | |
| 330 | 0.2 | 4.1 | | | | | |
| 360 | 0.15 | 2.9 | | | | | |
| 390 | 0.1 | 1.8 | 0.5 | 10.9 | 103 | 57.5 | -12 |

(iv) For comparison, a "stationary" test (without nitrogen flushing) was carried out in the same apparatus. After seven hours, the following set of figures was obtained :

$$(B) = 115 \quad (C) = 42.8 \quad (B-C) = 29.4$$

These experiments show that the elimination of HBr leads to the additional formation of hydrogen bromide, and finally to a negative (B-2C) value. The total amount of HBr formed in the stationary test is smaller, and (B-2C) is positive. From the operating conditions, it is clear that hydrolysis cannot be responsible for the HBr formation. Its cause can only be substitution or secondary decomposition of the primary addition product. The former alternative is excluded by the fact that (B) and (C) do not have a parallel increase : substitution would obviously demand such parallelism.

In contradistinction to Poell's assertion, it has thus been shown that at least the additional hydrogen bromide formed in the nitrogen experiments derives from the decomposition of a bromine addition product.

(c) Elimination of HBr by Shaking with Water

After bromination, the CCl₄ solution was separated, titrated as usual for Br₂ and HBr, and then shaken with aqueous KI solution until no more HBr was formed. Table VI shows that in this way the same results were obtained as in the experiments with nitrogen.

TABLE VI

Bitumen : 360 mg

Bromine solution : 15 ml, 26 mg per ml

| | Time of Reaction | (B) | (C) | (B-2C) |
|----|------------------|------|------|--------|
| 1. | 15 minutes | 85 | 42 | +1 |
| 2. | 30 minutes | 93.5 | 51.5 | -9.9 |
| 3. | 60 minutes | 98.3 | 54.0 | -9.7 |

The foregoing experiments point to the equilibrium nature of the reaction by which HBr is formed. It thus seems possible that bitumen is able to react with hydrogen bromide. Whilst a carbon tetrachloride solution of the material did not react with aqueous hydrogen bromide, it did absorb HBr when this was applied in the form of a solution in the same organic solvent.

TABLE VII

Absorption of HBr by Bitumen, both being dissolved in CCl₄

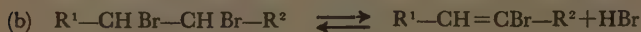
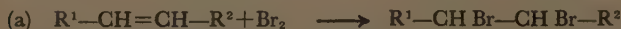
(Values in ml n/10)

Exp: (a) Addition of 10 ml HBr solution

Exp: (b) Addition of 20 ml HBr solution

| | HBr in ml n/10, after reaction | |
|---|--------------------------------|-----|
| | (a) | (b) |
| Blank : 10 ml CCl ₄ | 3.1 | 5.3 |
| Test : 360 mg bitumen in 10 ml CCl ₄ | 1.0 | 2.1 |
| Absorbed HBr by bitumen | 2.1 | 3.2 |

As it is well known that aliphatic double bonds add hydrogen bromide, this observation lends additional support to the above conclusion, which can be formulated as follows :



The true unsaturation value of bitumen is, therefore, (B), and no allowance need be made for the hydrogen bromide liberated. Bitumen contains unsaturated constituents, and Poell's experiments can now be interpreted in the following manner : During the distillation of the resulting solution, more and more hydrogen bromide was formed,

owing to decomposition of the bromine addition product, until eventually, for each molecule of bromine absorbed, one molecule of HBr was obtained. The residue did not react with bromine — not because, as Poell assumed, it was saturated (and was therefore a substitution product), but because the more highly substituted bromine-olefine (see equation b) had become refractory to bromine addition. This is in accord with the general experience of organic chemistry.

The bitumen used throughout this work was found to have the following chemical and physical properties:

| | | | |
|--------------------------------|-----------------|--|----------------------|
| <i>Chemical Analysis</i> | | Ductility (at 25 deg. C) | Exceeding 110 cm. |
| Ash | 0.059% | Specific Gravity | 1.02 |
| Sulphur (S) | 5.3% | Loss on Heating (5 hrs. at 163 deg. C) | 0.3% |
| Soluble in CS ₂ | 99.7% | Penetration after loss on Heating (at 25 deg. C) | 67 |
| Peroxide | none | Ductility after Loss on Heating (at 25 deg. C) | Exceeding 110 cm. |
| Acidity | none | | |
| <i>Standard Tests*</i> | | Oliensis Spot Test | Homogeneous |
| Penetration (at 25 deg. cent.) | 79 | Hexane Resistance | 3 ml (15 increments) |
| Softening Point | 51.5 deg. cent. | | |

* The Standard Tests were carried out according to relevant British and American Specifications (See: Abraham, Asphalts and Allied Substances, Fifth Edition, 1945).

ECOLOGICAL INVESTIGATIONS IN PALESTINE — II* ON THE VEGETATION OF KURKAR HILLS

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As first pointed out by Eig (1931), Israel is the meeting place of three different phytogeographical regions: the Mediterranean, the Irano-Turanian and the Saharo-Sindian, (Fig. 1). In addition, there are many Sudano-Decanian enclaves inside the Saharo-Sindian territory wherever the local physiographical conditions are adequate.

Phytogeographical map of Palestine Modified after Zohary, 1947

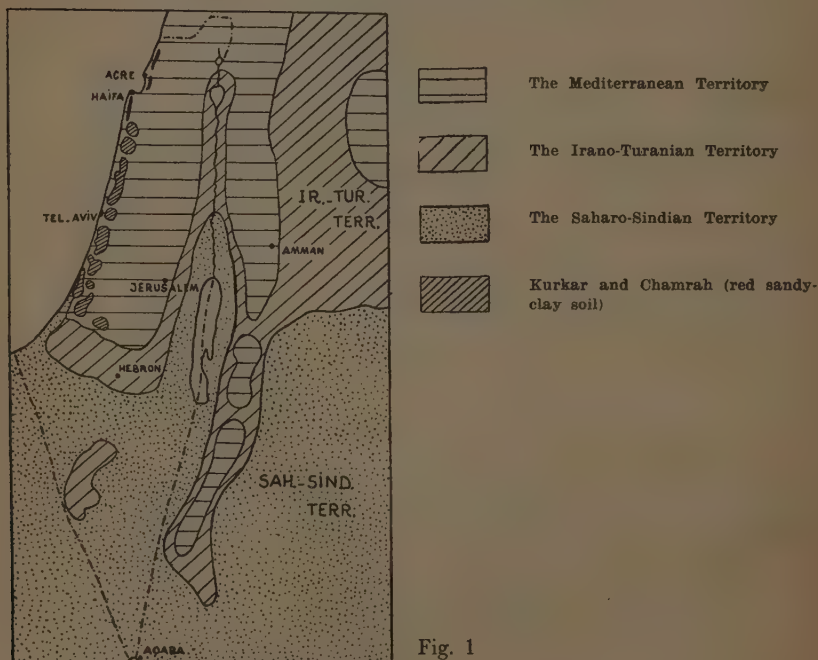


Fig. 1

The Mediterranean territory of Palestine is physiographically composed of two parts: the mountainous area (Central Judean hills, Upper Galilee, etc.) and the coastal plain.

* Part I appeared in *Pal. Jour. Bot. Jer. Ser.* 3:138-150.

stretching from Gaza approximately to Haifa. Inside the coastal plain, forming discontinuous low chains, are situated the Kurkar hills, which are most interesting from a geological as well as from a phytogeographical-ecological point of view. Lying within the Mediterranean territory, these hills, and the shallow valleys and small plains in between, bear a vegetation distinctly different from that of the surrounding Mediterranean area.

The Kurkar hills are composed of calcareous sandstone and, according to Loewengart (1928) and Picard and Avnimelech (1937), represent old fossil dunes. The dune sand was cemented together by the capillary ascent of limestone solutions and calcareous covers and limestone concretions were formed. Underlying the Kurkar, red sandy soils are chiefly found. Through weathering they form the red sandy soils, called Chamrah, which are to be found among the Kurkar hills.

The most typical plant association of the Kurkar hills is the *Helianthemum elliptici*, extensively studied by Eig (1939). It is a Saharo-Sindian community, mainly composed of such Saharo-Sindian elements as *Helianthemum ellipticum* and *Retama Roetam*. As the plant cover is very discontinuous and the plants are typical desert plants, the vegetational physiognomy of these hills is that of a desert. The Chamrah soils, on the other hand, are characterised by an *Eragrostis bipinnata* — *Centaurea procurrens* association in which, here and there, grow isolated trees of *Zizyphus Spina Christi* and *Ficus Sycomorus*, two typical Sudano-Deccanian elements. The high grass, together with the scattered trees, gives to this landscape a definite resemblance to an African savannah. It is probable that this is more than just a physiognomical resemblance and that this association really represents the most northern penetration of the African savannah.

These two completely different associations, growing side by side and separated by a small transition zone of only a few yards in places, constitute an interesting ecological problem. This research was undertaken to answer the following questions:

- (1) Why do Saharo-Sindian desert-like and Sudano-Deccanian savannah-like associations occur inside the Mediterranean territory under typical Mediterranean climatic conditions?
- (2) What ecological factors are responsible for the fact that two completely different associations, composed of different elements, grow side by side?

Description of Locality and Methods

In order to find an answer to these questions we chose two typical habitats in the vicinity of Ramat Gan, a town 8 miles north of Tel Aviv on the road to Haifa (Fig. 1). One habitat (Station I) was situated on the south-eastern slope of a typical Kurkar hill inside a typical *Helianthemum*. Station II was located inside an *Eragrostis* — *Centaurea* association in a small valley with characteristic red Chamrah soil more than

10 m. deep at the foot of the hill on which Station I was situated. A big and beautiful *Ficus Sycomorus* and a *Zizyphus Spina Christi* tree grew there. At a distance of 60 m. there was an orange plantation. We may mention here that the Chamrah soil in Israel is ideal for citrus plantations and *Eragrostetum* is the best indicator of the suitability of a given locality for citrus groves.

The plants chosen for the investigation were: Station I:—*Helianthemum ellipticum* (Desf.) Pers., *Retama Roetam* (Forsk.) Webb; Station II:—*Eragrostis bipinnata* (L.) Muschl., *Zizyphus Spina Christi* (L.) Willd., *Ficus Sycomorus* L.

During 1939/40 we measured a complete yearly cycle of osmotic values of plants (cryoscopic method) at the two stations; suction force of soils (Hansen's method, modified by Stocker [1930]); water content of soils and their chemical and physical composition.

Hansen's method has a drawback. When the soils are saturated with water the values measured are too small because there is no way of preventing the water vapour from condensing on the walls of the vessels and on the strips of filter paper when the vessels are taken out of the thermostat for weighing. In our tables the values obtained under these conditions are marked with an asterisk.

Osmotic Values of Plants and Suction Forces of Soils

In Israel, generally, the lack of water is by far the most important limiting factor for vegetation; hence we thought that it would be responsible for the occurrence of the desert vegetation on the Kurkar hills. As the water balance of a certain habitat is best expressed by osmotic values of plants and suction forces of soils (see Gradmann 1929), a study of Table 1 and 2 and Fig. 2, in which these values are presented, should indicate whether this supposition is correct.

The outstanding features of the osmotic values at both localities can be summed up as follows:

- (1) The two highest osmotic values measured occur in the *Eragrostetum* association. (*Eragrostis* 22. 84, *Zizyphus* 22. 36).
- (2) The greatest fluctuations in values are found in the *Eragrostetum*. The course of the curves in the *Helianthemetum* is much more even and stabilised. This is clearly seen from Fig. 2.

Fig. 2. — (a) Osmotic values of *Helianthemum ellipticum* (— — — —) and *Retama Roetam* (—) in atm. during the course of one year. (b) Osmotic values of *Eragrostis bipinnata* (—), *Ficus Sycomorus* (.....) and *Zizyphus Spina Christi* (— — — —) in atm. during the course of one year. (c) Rainfall for Ramat Gan in mm. for the different months of one year. (d) Suction forces of soils in *Helianthemetum elliptici* (—) and *Eragrostetum bipinnatae* (— — — —) in atm. during the course of one year in 60 cm. of depth.

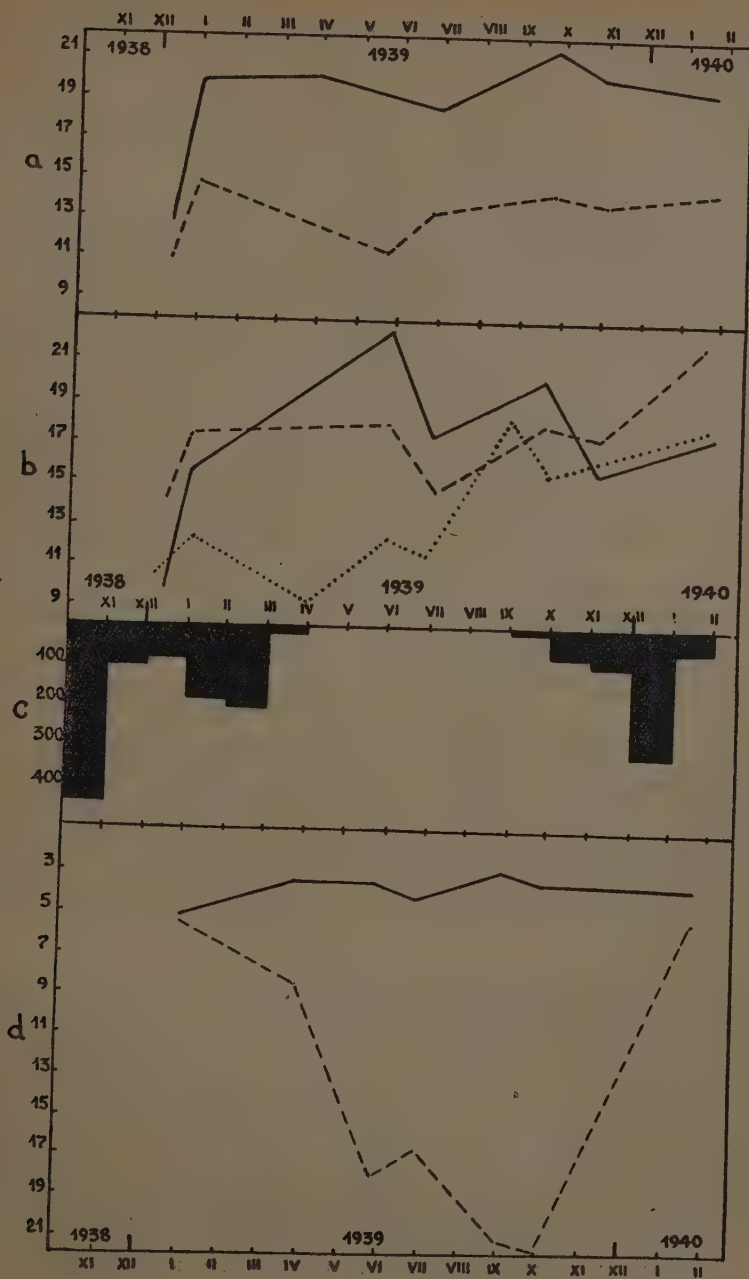


Fig. 2.

(3) In all plants there is a rise of values after the heavy rainfalls of the season 1938/39 which is followed by a fall later or earlier during the dry season. Then the curves rise again.

When the suction forces of the soils are considered (Table 1) there is a marked difference between the two stations which is much more pronounced than the differences in the course of the osmotic values. We may state the following :

(1) The values in all depths measured are higher in the *Eragrostetum* than in the *Helianthemetum*. This difference is most pronounced in the deeper soil layers where e.g. in 60 cm the suction force for the *Eragrostetum* rises during summer to 21.0 atm. whereas the highest value for the *Helianthemetum* in the same layer is only 5.2 atm. The respective values for 40 cm are 96.4 and 5.2.

(2) A comparison of the yearly course of the suction force values shows clearly (Fig. 2) that there is a definite rise of values in the *Eragrostetum* during summer which is missing in the *Helianthemetum*. This indicates that the water conditions of plants, as expressed in the soil suction forces, are much better at Station I than at Station II. The same can be seen in comparing osmotic values with suction forces. In the *Helianthemetum* we find that at depths of 40 and 60 cm, which is the main rooting level, the osmotic values during the whole year are much higher than the soil suction forces. The big difference between these two values is best shown if the highest suction force measured during the year is expressed in percentages of the lowest osmotic value. The figures are : *Helianthemum* 47.5%, *Retama* 40.3%.

In the *Eragrostetum*, conditions are completely different. At a depth of 40 cm the suction forces during the dry season are much higher than the osmotic values of any of the plants. Even at 60 cm the suction force is sometimes higher than the osmotic values. The difference between the two figures is never great. We had to dig to 80 cm and more in order to find lower suction forces during summer. The *Eragrostetum* plants can only maintain their water balance by sending their roots deep down into the soil. This is possible as, the depth of the red Chamrah soils is considerable, by comparison with most other soils in Israel.

When we compare osmotic values with the quantity of rain which fell during the investigation period, we understand why the osmotic values were higher during the rainy season of 1939/40 than during that of 1938/39. In 1938/39 our stations received 947.9 mm of rain; in 1939/40, only 589.6 mm.

But this comparison does not suggest why all the osmotic values show a definite descent during the dry season when, according to all other factors, we should expect a continuous rise, at least in the *Eragrostetum*.

This falling off is explained when we consider the phenology of the plants investigated. *Eragrostis*, *Ficus*, and *Zizyphus* formed new leaves in June which in July were already large and were taken for the determination of the osmotic values. It is well

known [Walter (1931), Thren (1933/34), Haas and Halma (1931), Oppenheimer and Mendel (1938) (for Citrus)] that the osmotic values in most plants decline during sprouting and that the osmotic value of newly formed leaves is lower than that of old ones. This was observed in the *Helianthemum* also, but there the new growth is formed much earlier.

There is another noteworthy point in the behaviour of the *Eragrostetum* plants. After the initial fall of the osmotic values in the beginning of the rainy season there occurs in all plants a considerable rise coinciding with the greatest seasonal quantity of rain, i.e., from December to February. Simultaneously the suction forces fall sharply. This seemingly strange behaviour is explained by the fact that at this time the leaves are in poor condition and appear dry and burned up. It is well known that leaves show much higher osmotic values before drying up and dying than during their active period. But why are these plants in poor condition although the *Helianthemum* plants are not? January and February are the coldest months in Israel with the lowest average temperatures of the year and the lowest average and absolute minima. Even though the temperatures in the coastal plain very rarely fall below zero during this period, apparently these plants suffer from cold, for at least two of them (*Ficus* and *Zizyphus*), being Sudano-Deccanian elements, are native to a much hotter climate. It is of interest in this connection that Haas and Halma (1931) for California, and Oppenheimer and Mendel for Israel, report for Citrus leaves "a definite trend towards higher osmotic values in winter when soil and air temperatures are low".

In summing up, we may say that poor water conditions cannot be held responsible for the occurrence of the desert vegetation on the Kurkar hills. The comparatively low osmotic values, their stable course throughout the year, and the low suction forces of the soils allow us to draw this conclusion. Comparison of the two stations and their plants shows clearly that the water relations in the *Eragrostetum* are definitely worse than in the desert-like *Helianthemum*.

Composition of the Soils

Having eliminated the water factor we investigated the other edaphic factors.

The soil of Station I is, as stated before, composed of typical Kurkar, i.e., sand containing large and small limestone concretions. On the surface, these concretions form an almost continuous uninterrupted layer. This "Panzer" layer is responsible for the fact that the Kurkar soils hold their humidity so well even during the hot summer months, for it protects the underlying soil from losing its water content by evaporation. The relatively high water content during the whole year at depths of 20 and 40 cm can only be explained in this way. Layers of pure sand without limestone concretions occurred only at a depth of a metre and more.

On Station II the soil is typical red Chamrah.

Table 3 contains the results of chemical and physical analyses of our soils. The figures are typical examples taken from a great number of analyses made for the two soils. The following conclusions may be drawn :

(1) The pH of the soils of the two stations is identical.

(2) The water content of air-dry soil is much greater in the *Eragrostetum* than in the *Helianthemetum*.

(3) Both soils are basically sandy as they contain between 92.0% and 97.37% of coarse and fine sand.

The soils of the *Helianthemetum* are 3—4% richer in sand than those of the *Eragrostetum*.

(4) There is a definite constant difference between the two types of soil with regard to their silt and clay content. The *Eragrostetum* soils are 2—3 times richer in these fractions than the *Helianthemetum*.

(5) The soil of either station is rich in water-soluble salts, but in this respect the *Eragrostetum* is always the richer of the two. Though the difference is not very great, it is constant. Both soils are poor in chlorides and completely lack soluble sulphates.

(6) The *Eragrostetum* soils are richer in nitrogen. In agricultural practice, soils containing less than 0.02% of total nitrogen are considered too poor for cultivation, from 0.02—0.03% as very poor, 0.03—0.06% as poor, 0.06—0.1% as moderately good, 0.1—0.2%, as good, 0.2—0.3%, as rich, and above that, as very rich. Various authors differ slightly as to these evaluations.

Classifying our soils on this basis, we find that all the soils of the *Helianthemetum* are either below the 0.02% limit or in the very poor category. All the *Eragrostetum* soils are in the poor or moderately good class.

(7) As to CaCO_3 content, the *Eragrostetum* soils are the poorer of the two but never show a complete lack of CaCO_3 . Eig (1939) and Zohary (1944) indicate an ab-

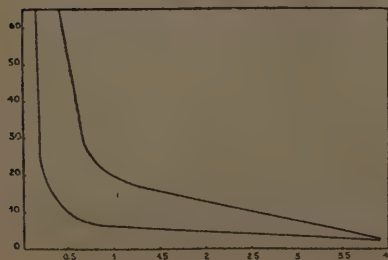


Fig. 3. — Suction forces of soils in relation to water content. 0-60 = Suction forces in atm. 0-40 = Water content in percentages of air-dry soils. Upper line = *Eragrostetum*; lower line = *Helianthemetum*.

soluble absence of CaCO_3 for the *Eragrostetum* soils. This contradiction may find its explanation in the fact that Station II, purposely selected near Station I, may contain limestone carried from the Kurkar hills into the adjacent red Chamrah soils.

The facts indicated in point 4 explain another phenomenon which we observed. The suction forces of the two kinds of soil were measured in relation to differing water contents. The results of these measurements are illustrated by Fig. 3.

When both contain the same quantity of water, the suction force in the *Eragrostetum* soil is higher than that in the *Helianthemetum*. As the two types of soil do not differ sufficiently in their salt content, this, as well as the fact that, when air-dry, the *Eragrostetum* soils contain more water than the soils of the *Helianthemetum* (see Table 3), can be explained only by the higher percentage of silt and clay of the *Eragrostetum* soils. This supposition accords well with the work of Oppenheimer and Elze (1941) who, working with similar soils of orange groves, found "a marked correlation between the percentage of clay and silt on the one hand and their water content at any given suction force on the other hand". Both figures and those of Mendel (1945) correspond well with our own data. Menchikovsky and Adler (1930), discussing the same red sandy clays, report that their water content when air-dry varied in accordance with the percentage of silt and clay.

Conclusions

The relatively high and very fluctuating osmotic values of the *Eragrostetum* plants, as well as the high suction forces of the *Eragrostetum* soil, show that these soils have, from a vegetational point of view, poorer water balance than the *Helianthemetum* soils. This is confirmed by a comparison of the suction forces of the two types of soil when both contain the same quantity of water. Under these conditions, the suction forces of the *Eragrostetum* are always higher than those of the *Helianthemetum* soils. The investigation excludes the possibility that the presence of the desertic *Helianthemetum* association on the Kurkar hills can be explained by bad water conditions.

The most probable explanation may be found in the fact that the *Helianthemetum* soils are poor in water soluble salts and, what seems to be more important, in nitrogen. The Kurkar hills would, therefore, represent a nitrogen, or perhaps a nutrient-element, desert and not a water desert.

This conclusion is supported by the observation that lack of nitrogen is typical of desert soils. "Desert plants must be adapted not only to very poor water conditions and high soil and air temperatures but also to a very low N-content and deficiency in other nutrient elements" (Evenari and Orshansky 1948 p. 11).

Though we can explain in this way the presence of Saharo-Sindian elements and associations inside the Mediterranean territory, as well as the fact that two completely

different associations grow side by side, there still remains the question of why Sudanian-Decanian elements are to be found on the Chamrah soils. To this our investigation offers no answer.

TABLE 1
Suction forces of soils in atmospheres

| Plant association | Depth in cm. | Date | | | | | | | |
|-------------------|-----------------|------|------|-------|-------|--------|-------|------|-------|
| | | 5.I | 30.I | 27.IV | 24.VI | 27.VII | 29.IX | 22.X | 16.II |
| Helianthemum | 0 | — | 5.2 | 249 | 249 | 249 | 249 | 249 | 6.6 |
| | 20 | 5.2 | 4.0 | 2.4 | 2.2 | — | — | — | 3.6 |
| | 40 | 2.4* | 5.2 | 3.6 | 4.0 | 4.0 | 3.3 | 4.6 | 3.3 |
| | 60 | — | 5.2 | 3.3 | 3.6 | 4.2 | 2.8* | 3.3 | 3.6 |
| | 80 | — | — | — | — | — | 2.4* | 3.6 | 2.8* |
| Eragrostetum | 0 | 4.6 | 21.0 | 207 | 249 | 249 | 249 | 249 | — |
| | 20 | 3.3 | 5.6 | 10.5 | 207 | 249 | 249 | — | 4.3 |
| | 40 | 5.6 | 4.0 | 5.3 | 96.4 | 96.4 | 69.8 | — | 4.5 |
| | 60 | — | 5.3 | 8.4 | 17.8 | 16.6 | 21.0 | 21.5 | 5.3 |
| | 80 | — | — | — | — | — | — | 12.5 | 4.3 |

TABLE 2
Osmotic values of plants in atmospheres

| Name of plant | Date | | | | | | | | |
|---------------|-------|-------|-------|-------|--------|-------|-------|-------|-------|
| | 5.I | 30.I | 27.IV | 24.VI | 27.VII | 29.IX | 22.X | 30.XI | 16.II |
| Helianthemum | 10.96 | 14.80 | — | 11.32 | 13.48 | — | 14.44 | 14.20 | 14.56 |
| Retama | 12.88 | 19.72 | 20.12 | 19.00 | 18.76 | — | 21.64 | 20.20 | 19.48 |
| Zizyphus | 14.56 | 17.56 | — | 18.04 | 14.68 | — | 18.04 | 17.56 | 22.36 |
| Ficus | 10.24 | 12.16 | 8.67 | 12.04 | 10.84 | 18.16 | 14.92 | 15.88 | 17.20 |
| Eragrostis | 9.63 | 15.40 | — | 22.84 | 17.32 | — | 20.44 | 15.88 | 17.92 |

* Percentages of air-dry soil.

TABLE 3

| Analysis | Helianthemetum | | | | Eragrostetum | | | |
|------------------------------------|----------------|---------|---------|---------|--------------|---------|---------|---------|
| | Depth in cm. | | | | Depth in cm. | | | |
| | 0—20 | 20—40 | 40—60 | 60—80 | 0—20 | 20—40 | 40—60 | 60—80 |
| H ₂ O in air-dry soil* | 0.33 | 0.34 | 0.17 | 0.19 | 0.61 | 0.81 | 0.58 | 0.64 |
| CaCO ₃ in air-dry soil* | 5.8 | 9.4 | 8.1 | 10.4 | 7.4 | 6.5 | 7.8 | 8.0 |
| pH | 7.4 | 7.4 | 7.4 | 7.4 | 7.4 | 7.2 | 7.4 | 7.4 |
| <i>Water extract *</i> | | | | | | | | |
| HCO ₃ | 0.067 | 0.067 | 0.059 | 0.067 | 0.084 | 0.062 | 0.084 | 0.113 |
| Cl | 0.004 | 0.006 | 0.005 | 0.006 | 0.003 | 0.003 | 0.006 | 0.002 |
| SO ₄ | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| Total salts | 0.016 | 0.016 | 0.048 | 0.304 | 0.032 | 0.024 | 0.504 | 0.416 |
| <i>Mechanical analysis</i> | | | | | | | | |
| Diameter of grains in mm. | | | | | | | | |
| 2.00—0.25 | 10.27 | 16.96 | 15.50 | 18.30 | 14.04 | 20.50 | 22.10 | 16.70 |
| 0.25—0.02 | 87.10 | 80.04 | 80.38 | 78.46 | 79.08 | 72.62 | 71.29 | 77.43 |
| 0.02—0.002 | 1.38 | 1.50 | 1.62 | 1.60 | 4.00 | 3.63 | 3.37 | 2.25 |
| below 0.002 | 1.25 | 1.50 | 1.50 | 1.62 | 2.88 | 3.25 | 3.24 | 3.62 |
| <i>N content</i> | | | | | | | | |
| Total N* | 0.00792 | 0.00899 | 0.03586 | 0.02376 | 0.02975 | 0.06319 | 0.07307 | 0.05504 |
| Organic N** | 95.71 | 93.00 | 97.24 | 94.91 | 95.63 | 91.65 | 96.84 | 98.38 |
| Inorganic N** | 4.29 | 7.00 | 2.76 | 5.09 | 4.37 | 8.35 | 3.16 | 1.79 |
| Ammonia N** | 3.03 | 4.45 | 1.08 | 1.85 | 2.65 | 1.41 | 1.07 | 0.88 |
| Nitrate and Nitrite N** | 1.26 | 2.55 | 1.68 | 3.24 | 1.72 | 6.94 | 2.08 | 0.91 |

*Percentages of air-dry soil.

**Percentages of total N.

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EGG-DEVELOPMENT AND DIAPAUSE IN THE MOROCCAN LOCUST

(*Dociostaurus maroccanus* Thnb.)

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In the embryological development of the Moroccan locust four stages are described, three of which represent various manifestations of the diapause phenomena. The relation of these four stages to the normal annual climatic cycle of the locust areas is discussed. No premature breaking of any of these diapauses could be achieved by artificial means. Several hitherto unexplained phenomena observed in reservations of the Moroccan locust are interpreted on a physiological basis.

INTRODUCTION

Locusts and grasshoppers show a great range of manifestations of the phenomena known as diapause, which appears, as a rule, in the egg stage. For the purpose of the present paper, we define diapause as any temporary retardation or temporary interruption of development. This paper will add, to the great range of diapause-phenomena in locusts, another complicated case. For information on the general life cycle and the general ecology of the Moroccan locust, the reader is referred to the work of Uvarov⁷ and Bodenheimer¹. Our experiments lasted from 1942 to 1947, when we received the last parcel of eggs from the Iraqi Directorate General of Agriculture. In the egg-development of *Dociostaurus* we have established four different phases, three of which can claim to be called diapause although each represents another physiological type of diapause.

MORPHOGENESIS AND BLASTOKINESIS OF THE EGG

Before entering into any discussion of diapause, we must study the morphology of egg-development. Only by assigning the proper stage of development to the embryos of the eggs dissected month by month is it possible to separate clearly periods of active development from those of interrupted development. No extraordinary observations were

made on the morphogenesis of *Dociostaurus maroccanus*. We therefore describe and represent briefly the twenty stages of development which we distinguish. The first fourteen differ from each other with regard to morphological characteristics; the last six mainly with regard to size and position within the egg.

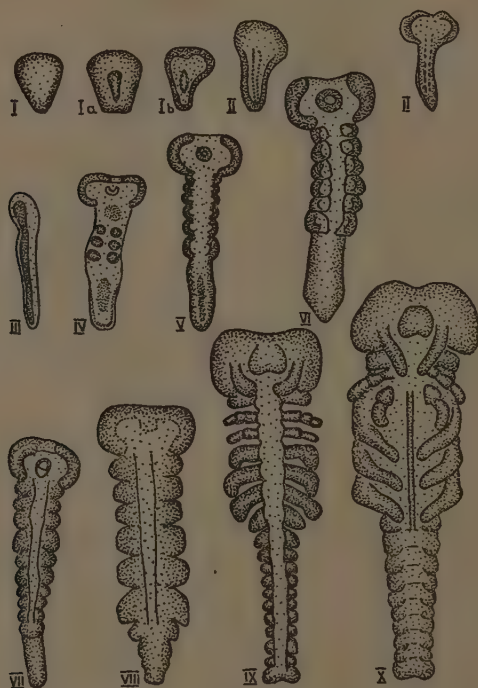


Fig. 1a. — Morphogenesis of stage I to X of the embryo of *Dociostaurus maroccanus*

These latter comprise the katatrepsic stages.

Stage I: In its initial stage, the embryo is short and triangular, with rounded corners, bearing no marks of differentiation. It is located in the neighbourhood of the micropyle and is 0.15 mm. long. With the beginning of development, the hind margin becomes plainly blunted, while the central groove appears. During these early changes the embryo migrates slowly ventrad. The broad cephalic end extends laterad and thereby forms the protocephalic lobes. The oral groove becomes deeper and more elongated.

Stage II: The body caudad of the protocephalic lobes, which continue to increase in width, stretches along its longitudinal axis, the caudal end now being well-rounded.

The central groove extends as a dorso-median furrow from the hind end of the protocephalic lobe to the caudal end of the embryo. Total length, 0.2 mm.

Stage III: The main feature of this stage is the growth along the longitudinal axis; this growth is twice that of *Stage II* and reaches a length of 0.4 mm.

Stage IV: The first signs of appendages make their appearance: the antennae and the three pairs of gnathal appendages (mandibles and maxillae). The labrum, also, is now formed. Eye-buds can be distinguished at the lateral parts of the protocephalic lobes. Length, 0.5 mm.

Stage V: Longitudinal growth continues. The labrum is distinctly marked. In addition to the cephalic appendices, those of the thorax also make their appearance. Length, 0.6 mm.

Stage VI: In addition to the continued longitudinal growth, a marked transversal growth begins. The thoracic extremities are now conspicuously more developed than the cephalic ones. Length, 0.7 mm.

Stage VII: The extremities of the first abdominal segments appear. The migration of the embryo toward the anterior end is now much advanced. Length, 0.9 mm.

Stage VIII: The leg-buds continue to appear on all abdominal segments, while the antennae begin to take a definite shape. The embryo reaches its temporary resting location anterad, at $\frac{3}{4}$ of the total egg length, where it remains until the start of katatrepsis. Length, 1.0 mm.

Stage IX: The antennae grow and extend caudad towards the mandibles. The mouth-parts are definitely shaped. All abdominal segments bear leg-buds. Length, 1.5 mm.

Stage X: The thoracic legs and the abdominal leg-buds fold ventro-mediad. Thorax and abdomen increase in width. Length, 1.9 mm.

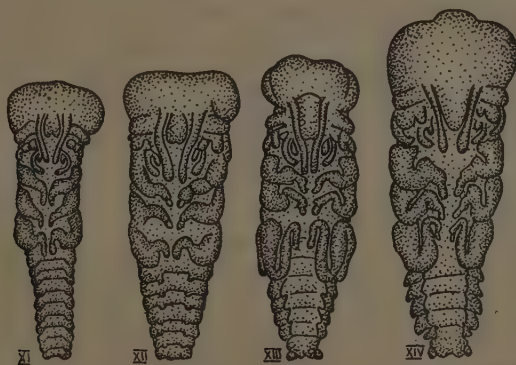


Fig. 1b. — Morphogenesis of stages XI to XIV of the embryo of *D. maroccanus*

Stage XI: Further growth and differentiation of cephalic and thoracic appendages. Labrum with distinct indentation at the caudad end. The maxillary palpi become dis-

tinct. Bends in the legs indicate the location of the future articulations. Length, 2.1 mm.

Stage XII: Conspicuous transversal growth to almost twice its earlier width. The optical lobes show considerable development. On the thoracic legs the tibio-tarsal articulations develop. The abdominal leg-buds are reduced to short stumps. Length, 2.3 mm.

Stage XIII: The antennae grow to reach the forelegs. Tibiae and femora of the thorax grow in length and assume their definite shape. The abdominal leg-buds disappear entirely. Length, 2.7 mm.

Stage XIV: The head shows preferential growth in length and width, and therewith the morpho-differentiation is at its end. The embryo now rests at the posterior end of the concave surface and extends over one third of the total egg length. The anatrepsic migrations of the embryo within the egg are ended. The coming katatrepsic phase is not connected with any further morpho-differentiation, but is characterized only by increase in size and is a period of heavy migrations.

Stage XV: The embryo begins slowly to migrate toward the convex side of the egg. Its width increases.

Stage XVI: The posterior curvature of the egg is almost filled. With strongly bent longitudinal body-axis, the embryo passes along the transversal axis of the egg. Continued longitudinal and transversal growth.

Stage XVII: The embryo has by now almost completed its migration to the convex side; only the caudal end of the abdomen still remains on the concave side.

Stage XVIII: The migration across the transverse axis is completed. The embryo's head is close to the anterior end of the egg and it now turns along its longitudinal axis; its venter, turning toward the concave egg surface, is now facing laterad.

Stage XIX: Migration along the longitudinal axis is completed, although it is still growing in length and width. The yolk is reduced to small relics. The dorsal ends of both sides meet and begin to close in the later larval dorsal mediane.

Stage XX: Katatrepsis is completed. The head of the embryo has reached the anterior end of the egg and the last relics of yolk have disappeared.

Fig. 2 illustrates the major phases of ana- and katatrepsis. The longitudinal cross-section is in the mediane of the upper and lower (*i.e.*, the concave and convex) surfaces of the egg. The transversal cross-section has always been made at the point of the embryo's greatest body width. Attention is called to Stage XVIII, in which the laterad return of the embryo from the convex to the concave egg-surface is conspicuous, in contrast to all other blastokinetic movements which take place almost entirely over the longitudinal axis of the egg. Apparently, however, this latero-ventrad migration can occasionally be absent.

It is obvious from the above that our stages are entirely arbitrary divisions of a continuous slow transformation, differentiation, and growth. Yet the designation of arbitrary stages is necessary in order to understand our further references to certain

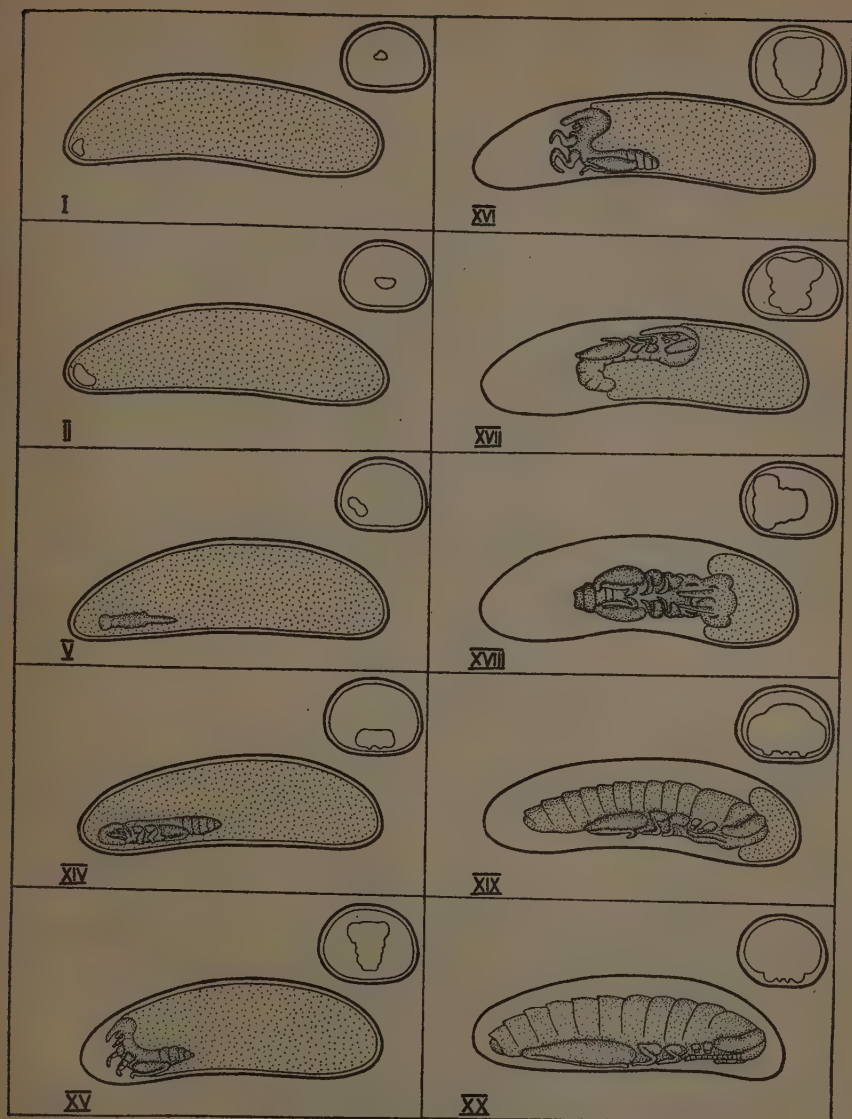


Fig. 2

Embryo of *Dociostaurus*, in situ in the egg, in Stages I, II, V, and XIV through XX, to illustrate growth and blastokinesis. The added cross-section of the egg is taken from the largest part of the embryo.

stages of development. We could as well have designated ten or seventy-five different stages in the development of *Dociostaurus maroccanus*; in fact we have chosen only the most conspicuous.

ANNUAL CYCLE OF *D. MAROCCANUS* EGGS IN NATURE

Normal annual cycle. In the Mosul area, larval hatching occurs usually in March-April. In 1943, when hatching was delayed by a cool late winter, the first larvae hatched around the 5th of April. The first adults appeared the 8th of May. The main oviposition was in the third decade of May. After oviposition in a bare, hard dry soil, the egg-stage lasts until hatching in the following spring, (Bodenheimer¹).

Stage I of the embryo appears on the third day after oviposition. Development may then continue immediately or be interrupted for one to three months. We were unable to discover any environmental factor which decides this difference in behaviour. The development of the embryo from Stage I to Stage IX also shows a great range of variation and is less synchronous than any later phase. The majority of the eggs reach the early polypodous stage (IX) in the second month after oviposition, many going on, during this month, to Stage XII. The third month brings the bulk of the eggs up to the oligopodous Stage XIII.

TABLE I.
Development of 62 eggs.
(No. of eggs reaching each stage)

| Days after oviposition | O | I | II | III | S IV | T V | A VI | G VII | E VIII | IX | X | XI | XII | XIII |
|---------------------------|---|----|----|-----|---------|--------|---------|----------|-----------|----|---|----|-----|------|
| 0-30 | 1 | 12 | 3 | | | 1 | 1 | | | | | | | |
| 31-60 | | 1 | 1 | | | 2 | 3 | 1 | | 4 | 9 | 4 | 8 | |
| 61-90 | | 1 | | | 1 | | | 1 | | | | | | 8 |

From the end of the fifth month on, but usually a few months later, Stage XIV is reached by almost all eggs. This stage is best characterised by the relatively important growth of the cephalic region as well as by the final differentiation of the tarsal joints. From Stage XIII-XIV until twelve to fourteen months after oviposition, the eggs are ready to continue development as soon as adequate conditions are offered. These are first, the presence of contact water; and second, favourable temperatures above the threshold of development. In Iraq these conditions are found in the natural sites in late February or March. If no contact water has been available by the end of twelve to fourteen months after oviposition, the eggs dry up and no development takes place. These results are obtained either on the natural oviposition sites or in the laboratory at 27° and 75% R.H.

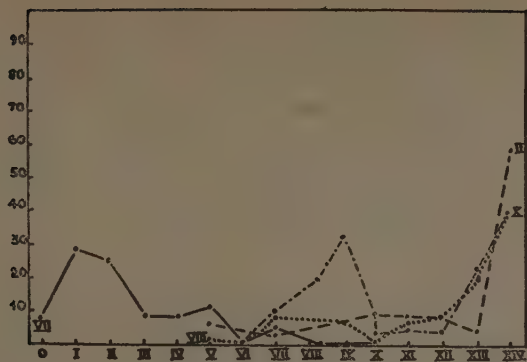


Fig. 3. — Percentages of eggs in various Stages (0-XIV) when kept at room-temperature and -humidity, during June, July, October and February.

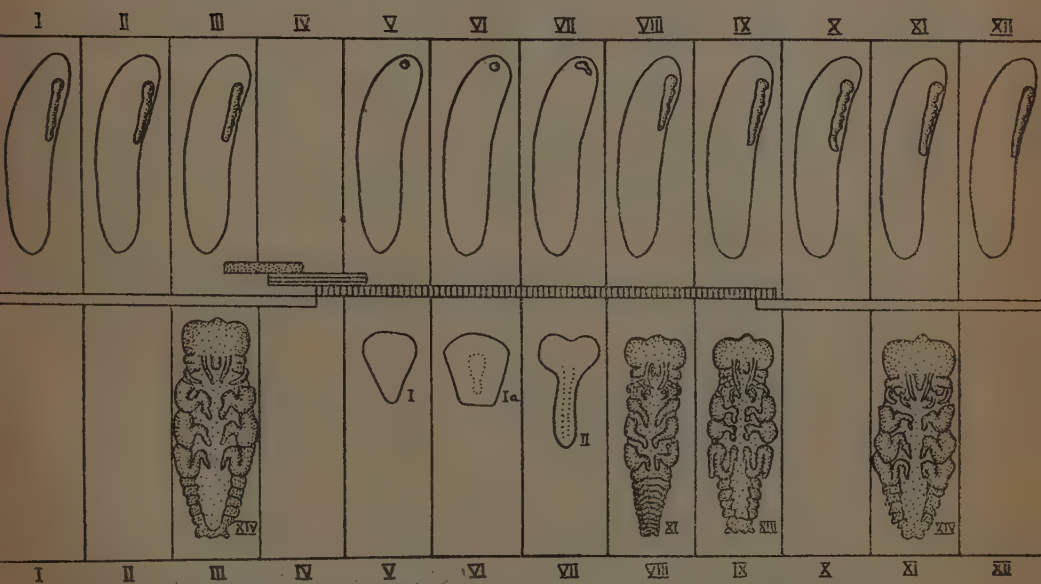


Fig. 4. — Egg development of *D. maroccanus*, month by month, in its natural environment.

Above: Size and position of embryo in egg.

Centre: Normal seasonal appearance of all stages:

Dotted lines: larval

Horizontal lines: adult

Vertical lines: eggs in phase A and B

White lines: eggs in phase C and D

Below: Stage of the embryo most common in every month.

We can thus divide the egg-development into the following phases :

A. Initial period. This extends from oviposition to the end of Stage I, which is reached within the first three days after oviposition. Then the embryo may either continue development and pass into phase *B*, or development may be interrupted for one or two months, occasionally even longer.

B. Anatrepsis. This phase includes development from Stage II to Stage XIII/XIV. While some eggs pass through this phase steadily, reaching this temporary goal within the second month after oviposition, other eggs, developing much more slowly, reach the same during the third to fifth month after oviposition.

C. Dormant period of readiness. Once Stage XIII/XIV is reached, development is interrupted but the eggs are ready to continue development — after an additional physiological maturation — at any moment when suitable environmental conditions are offered. This occurs on the natural sites in late winter of the following year. This means that five to eight months are passed, in nature, in this phase.

D. Katatrepsis and hatching. When embryos in Stage XIV become wet, they speedily begin katatrepsis. At the same time, they grow rapidly until the entire egg is filled and they are ready to hatch. The duration of this phase depends mainly upon the environmental temperatures.

From a theoretical point of view, it is difficult to decide which of these phases deserve the name of "diapause". When development stops for some time at Stage I, this certainly is diapause. Very slow and inhibited development, in phase *B* may also be called diapause. When lack of suitable environmental conditions, during Stage XIV, interrupts development for five months or longer, this interruption may also be termed diapause. Yet in each of these cases the physiological condition of diapause is entirely different. The only common symptom is an inhibition or interruption of development. It is obvious that the entire theoretical background of the problems connected with diapause deserves a critical revision; this is reserved for discussion elsewhere.

Eye pigmentation develops mainly during Stages XIII/XIV. Its appearance has been taken, by earlier field workers, as a sign of approaching hatching. Yet directed observation showed us that intensive black pigmentation of the eyes may appear as early as Stage VIII, very late in Stage XIII, or even later. It thus appears that eye pigmentation cannot be used as an accurate indicator of the stage of development.

The general trend of egg-development in *Dociostaurus maroccanus* resembles most nearly that of the Australian locust *Austroicetes cruciata* Sauss. (Steele⁶), from which it differs mainly in the facultative arrest of development in Stage I. It is also similar to the egg-development of *Melanoplus differentialis* Thos. (Carothers²) and of *M. mexicanus* Sauss. (Parker⁵), but it differs from both in that high temperature does not further development before Stage XIV is reached.

The eggs within one egg-pod are not invariably in the same stage of development.

This is especially so in the earlier stages. Eggs of the same egg-pod do not always hatch contemporaneously, but the morphological differences, in such case, are negligible.

For practical purposes we may state that below 15° no hatching takes place.

Changes in weight and specific gravity during development. Oviposition is followed by some loss of water from the egg. The average weight of the intra-abdominal egg, ready to be laid, is 6.0–7.0 mg. It drops within one to two weeks to 5.5–6.0 mg. When, in Stage XIII, it is ready to complete development upon suitable extero-stimulation, weight has dropped to 4.5–5.5 mg. Yet eggs weighing only 3.5 mg during the normal period of development remain viable.

With the imbibition of contact water, the weight suddenly almost doubles to 8.0–10.0 mg. The hatching vermiform larva has a weight of about 8.3 mg which is reduced, after shedding of the so-called amnion, to 7.0–8.0 mg.

TABLE II

Average weight and specific gravity of D. maroccanus eggs kept in dry soil or dry atmosphere

(Within egg-pods)

The figures represent averages of at least 50 eggs

| Month | Age | Embryo Stage | Wt in mg | Spec. Gravity | Eggs Dry Wt mg | % Total Egg Wt |
|-------|------------|--------------|----------|---------------|----------------|----------------|
| | 10 min | 0 | 7.0 | 1.100 | | |
| | ½ hr | 0 | 6.5 | 1.091 | | |
| | 1 hr | 0 | 6.5 | 1.091 | | |
| | 1-2 days | 0 | 5.6 | 1.108 | | |
| | 3-13 days | I-II | 5.9 | 1.108 | 2.3 | 50 |
| May | 16-26 days | I-VIII | 6.0 | 1.109 | | |
| June | 1 mo | I-IX | 5.5 | 1.100 | | |
| July | 2 mo | " | 5.5 | 1.097 | | |
| Aug. | 3 mo | II-XIII | 5.5 | 1.084 | | |
| Sept. | 4 mo | " | 5.0 | 1.096 | | |
| Oct. | 5 mo | " | 5.4 | 1.089 | 2.3 | 55 |
| Nov. | 6 mo | XIII-XIV | 5.5 | 1.100 | | |
| Dec. | 7 mo | " | 4.8 | 1.091 | | |
| Jan. | 8 mo | " | 4.6 | 1.093 | 1.9 | 53 |
| Feb. | 9 mo | " | 4.5 | 1.095 | | |
| Mar. | 10 mo | " | 4.8 | 1.097 | | |
| Apr. | 11 mo | " | 4.5 | 1.096 | 2.2 | 52 |
| May | 12 mo | " | 4.7 | 1.097 | | |
| June | 13 mo | " | 4.7 | 1.097 | | |

In the beginning, the specific gravity fluctuates around 1.100, dropping to about 1.085 and then rising to about 1.096 where it remains constant during the entire phase of readiness to hatch. Following the sudden imbibition of contact water, the specific gravity drops to 1.059—1.065 where it remains until the egg hatches.

Dr. B. Shapira kindly determined the fat content of eggs which were received in July and kept at room-temperature and -humidity.

| Fat Content | July | Aug | Sept | Oct | M | O | N | T | H | Jan | Feb | Mar | Apr |
|----------------|------|------|------|------|------|------|------|------|------|------|-----|-----|-----|
| % dry weight | 49.5 | 50.2 | 50.4 | 50.5 | 50.6 | 49.8 | 49.8 | 50.3 | 49.6 | 49.8 | | | |
| % fresh weight | 7.1 | 6.8 | 6.5 | 5.9 | 6.3 | 5.6 | 3.0 | 2.7 | 2.6 | 2.7 | | | |

After wetting, the eggs showed such marked fluctuations of fat-content that Dr. Shapira prefers to consider the results not reliable. At present we can only point out the large drop in fat-content of the egg in early January — about the time when the weight of the egg normally drops — which indicates physiological changes preparatory to initiation of phase *D* under natural conditions.

TABLE III

Changes in weight and specific gravity of eggs after wetting at 27°

| Embryo | | | Eggs | | |
|--------------------|-------------------------|---------|------------------|-----------|----------------|
| Days after wetting | Stage | Wt (mg) | Specific Gravity | Dry Wt mg | % Total egg wt |
| 1 | XIV | 5.4 | 1.100 | | |
| 2 | XV | 6.1 | 1.088 | | |
| 3 | | 7.7 | 1.077 | | |
| 4 | XVI | 8.2 | 1.069 | | |
| 5 | XVII | 8.3 | 1.065 | | |
| 6 | | 8.5 | 1.067 | | |
| 7 | XVIII | 8.6 | 1.064 | | |
| 8 | XIX | 8.6 | 1.064 | | |
| 9 | XX | 9.2 | 1.061 | | |
| 10 | | 8.7 | 1.065 | | |
| 11 | | 9.4 | 1.060 | | |
| 12 | Hatching | 9.2 | 1.059 | | |
| | Vermiform larva | 8.3 | | 1.6 | 24.0 |
| | After shedding "amnion" | 7.0 | | 2.6 | 21.3 |

Viability of eggs when wetted at various months. We have mentioned above that the eggs lose their viability when no wetting occurs until the following spring. Results of our experiments are condensed in Table IV. The eggs were kept, before wetting, at room temperature and after, at 27°.

TABLE IV

Correlation between wetting and degrees (0-3) of total development at various months.
(s indicates spoilage of egg)

| Year | Month of wetting | | | | | | | | | |
|------|------------------|-----|-----|-----|-----|-----|-----|-----|-----|------|
| | Sept | Oct | Nov | Dec | Jan | Feb | Mar | Apr | May | June |
| 1942 | | 0 | 1 | 2 | 3 | 2 | 2 | | 2 | |
| 1943 | | 0 | 2 | 2 | 2 | 2 | 3 | | 2 | |
| 1944 | | | | 2 | 2 | 2 | 1 | 3 | 2s | 0 |
| 1945 | 0 | 1 | 3 | | 3 | 3 | | 2s | 1s | |
| 1946 | | | 0 | 2 | 2 | 3 | 3 | 2 | 2 | 1 |

The loss of viability of the eggs is accompanied by some conspicuous changes. The yolk shows granulation and becomes foamy and brittle; the egg breaks easily. Sometimes the embryo and part of the yolk become black. This blackening spreads from the micropyle all over the egg. The discolouration begins in the embryo at the tarsi, but is most conspicuous in the head. Eggs kept in a dry atmosphere show this change beginning in May of the year following oviposition. When the egg, after oviposition, is punctured by a needle, similar blackening may set in four days after the puncturing. Removal of a small patch of the chorion also induces blackening two weeks later. Single eggs which are exposed, a short time after oviposition, to 40—70 or 100% R.H. also show blackening.

Enzymatic changes during egg-development. At the suggestion of the senior writer, Dr. N. Lichtenstein⁴ and Dr. P. Fodor⁵ studied the activity of proteolytic and lipolytic enzymes in phases *C* and *D* of the egg-stage of *D. maroccanus*. The proteases in phase *C* did not hydrolyse casein or gelatine, but readily hydrolysed casein peptone, gelatine peptone, leucoglycine, leucylglycylglycine, and chloroacetyl tyrosinase. In phase *D* definite cleavage of casein was obtained. The butyrine-cleaving and olive-oil splitting lipases did not show such basic difference in the two phases. It is intended to continue such enzymatic analyses as soon as new material of *Doclostaurus* eggs is available.

Influence of temperature on the duration of the post-diapause phase D. Hibernated egg-pods were wetted in spring and exposed to a series of different temperatures; ten pods were exposed at each temperature level. At 15—27° most of the eggs hatched; at 30°, a few; and at 33°, one egg only.

TABLE V
Duration of phase D after exposure to various temperatures

| Breeding Temperatures | 14.9 | 15.7 | 18.9 | 19.2 | 23.3 | 26.2 | 29.4 | 32.0 | 32.7°C |
|-----------------------|---------------------|------|------|------|------|------|------|------|--------|
| | Days after exposure | | | | | | | | |
| First larval hatching | 61 | 49 | | 25 | 16 | 12 | 9 | | 11 |
| Bulk larval hatching | 62 | 51 | 21 | 28 | 17 | 13 | 11 | 12 | |

Vermiform larvae hatching at temperatures below 14.5° rarely succeeded in shedding the "amnion" and usually died.

These, and other breedings, yield a temperature-development hyperbola with the following constants:

c. (Temperature-threshold of development)=12.1°

th.c. (Thermal-constant)=164 day-degrees.

The upper turning point of the hyperbola, *i.e.* the retardation of development at high temperatures, begins at about 32°.

EXPERIMENTS IN INFLUENCING NORMAL EGG DEVELOPMENT

I. TEMPERATURE

- (1) The following experiments were conducted, in November, on eggs in Stage XIII. Contact with water was available to them only as indicated below.

A. Experiments with egg-pods. (Ten egg-pods in each group).

- a. Eggs were cooled for fifteen to thirty minutes at low temperatures (-10°) and then distributed at 27° over various R.H.
- b. As in "a" except that the eggs were wetted for 24 hours following cooling.
- c. As in "a" except that the eggs were kept one week at low temperatures (8-10°).
- d. As in "c" except that the eggs were wetted for 24 hours after cooling.
- e. Eggs were first wetted without cooling and then put into temperatures of 15, 20, 25, 30, and 35° and R.H. of 70 and 40%.
- f. As in "e" without wetting.

B. Experiments with egg-pods. (Ten egg-pods in each group).

- g. Exposure to 40, 70, and 100% R.H.
- h. As in "g", after removal of a small patch of the chorion.
- i. As in "g" except that eggs were punctured until a droplet of fluid appeared.
- j. As in "g" except that eggs were initially wetted for 24 hours.

In none of the series a-j was katatrepsis observed, nor did any of these experimental groups develop normally when transported into suitable environment the following autumn. Development of phase D was not initiated in any of the groups where contact water was not available. Normal development was not initiated in any of those groups to whom a moderate amount of contact was available for only 24 hours.

(2) From July on, egg-pods were exposed every month to cooling and were subsequently placed for observation upon wet filter paper. In most series exactly the same results were obtained as in the control series. In one group only, in September, the morpho-physiological readiness was advanced by almost one month as compared with the control. It is doubtful whether this single exception can be regarded as significant.

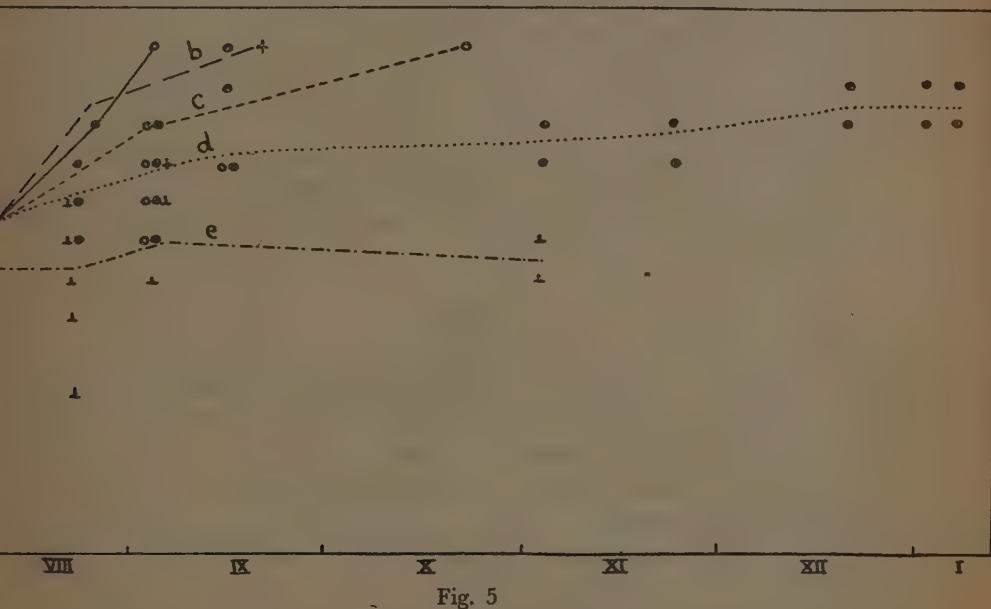


Fig. 5

Morphological development of embryos exposed at Stages VII-VIII in early August on dry soil, at 75% R.H., to various extremes of temperature.

"a": room temperature, 3 hrs daily to 8°; "b": at 8°; "c": at 33°; "d": at 35°; "e": at 40°.

It illustrates the fact, however, that under optimal conditions in autumn (which are not found, as a rule, in the normal habitats of the Moroccan locust) partial hatching can be induced.

(3) Influence of extreme temperatures upon the morphological differentiation of the eggs was shown when, in early August, egg-pods in which the embryos had reached Stage III—V (probably 30—45 days after oviposition) were exposed permanently in dry glass jars to the following temperatures and temperature alternations.

- At low (5—8°) temperatures, development reached Stage XIII, but with considerable delay.
- Embryos in egg-pods daily exposed to temperatures of 5—8° for three hours alternating with 21 hours at room temperature, developed at exactly the same

speed as the controls kept at room temperature; however, physiological readiness to pass into phase *D* appeared earlier in the experimental group.

- c. At 33° development was slightly retarded.
- d. At 35° development was inhibited and did not go farther than Stage XI or XII.
- e. At 40° only Stage X was reached.

At 35° and 40° the eggs dried up at an early stage. In series "a—c" the eggs hatched normally when later wetted and kept at 27°.

(4) Egg-pods of the same provenience as those in (3) were exposed to the same treatment two weeks later when the embryos were in Stages XI to (mainly) XIII. Only the eggs of "b" hatched later.

(5) The same treatment as in (4) was initiated eleven days later when all embryos were in Stage XIII. Those treated as in "a" and "b" remained alive and hatched eventually, while all those subjected to higher temperatures did not develop and became blackish in December.

II. HUMIDITY

(1) Immediately after oviposition eggs were placed in thoroughly wet soil. (Whenever the soil remains very wet for a few days, all eggs die before reaching Stage I.) When egg-pods (three in each group) were transferred from wet soil into different percentages of air humidity, development proceeded as follows:

- 100 About 10% of the eggs developed to Stage XIII, when 90% of these died. Others were killed by moulds.
- 90 A very few, by chance, reached Stage XI. The others died earlier.
- 80 Most of the eggs reached Stage XIII.
- 70 Most of the eggs reached Stage XIII.

At lower humidities, anatrepsis was not completed. Teratological embryos occurred in Stage VIII at a R.H. of 60%. At 10% R.H., in one group only, embryos reached Stage II; the others dried up during the first month.

In wet soil which was drying up, slow development was observed until Stage XIII. When the egg-pods were exposed alternately to wetting and drying, no development took place.

(2) Influence of R.H. on phase *C* and *D* was demonstrated when eggs in Stage XIII were allowed 7 days in contact water, and then restored to their original R.H. Development was completed when conditions of high R.H. (90 and 100%) and permanent availability of contact water were maintained until the end of development. At 80% R.H., and lower, no hatching took place, even if the eggs were transferred into 100% R.H. for the final period. At 100% R.H., normal hatching occurred only when an adequate amount was imbibed at the beginning.

The results concerning phase *D* are compiled in Table VI.

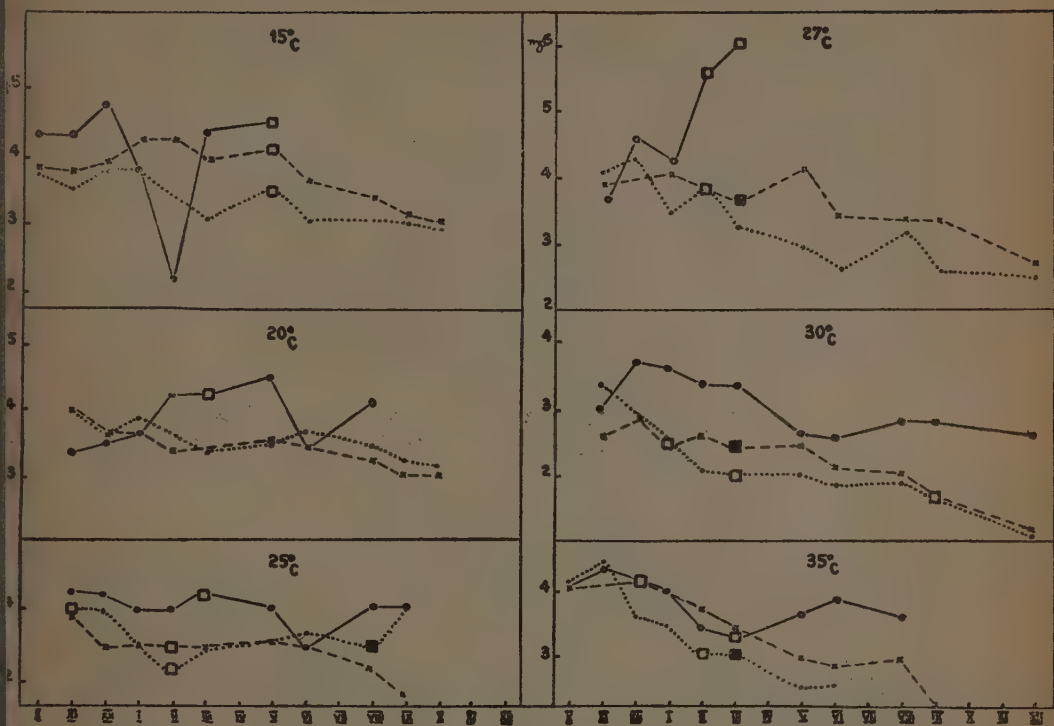


Fig. 6. — Change in weight of *D. maroccanus* eggs, in phase C, Stage XIV, at various temperatures and humidities

Dotted line: 0-40% R.H.; interrupted line: 60-80% R.H.; full line: 100% R.H.

TABLE VI

*Correlation between R.H. and development (weight of egg) in phase D.
(at 27°)*

| Date | 40% R.H. | | | 60% R.H. | | | | 100% R.H. | | | |
|--------|----------|-----|--------|----------|-----|-----|--------|-----------|-----|-----|--------|
| | C.W. | R1 | R2 | C.W. | R1 | R2 | R3 | C.W. | R1 | R2 | R3 |
| Jan 24 | 4.3 | | | 3.3 | | | | | | | |
| Jan 31 | 7.8 | 7.8 | | 5.8 | 5.8 | | | 3.6 | | | |
| May 2 | 8.1 | 2.5 | 8.1 | 8.0 | 5.5 | 8.0 | | 8.1 | 8.1 | | |
| Aug 2 | 8.0(H) | (d) | 5.0(d) | 9.0 | 6.0 | 6.0 | 9.0 | 9.1 | 8.1 | 9.0 | |
| Nov 2 | | | | (H) | 3.3 | 3.0 | 2.3(d) | (H) | 8.1 | 8.1 | 8.1(H) |

Date: given contact water

C.W.: contact water (permanent).

R1 : 1 week in C.W.

R2 : 2 weeks in C.W.

R3 : 3 weeks in C.W.

H : Hatched

d : died

III. OTHER EXPERIMENTS

Eggs which were totally submerged for some time (5 to 7 days) during any active stage of development, never developed. Part of them decayed and turned blackish, while others, though not showing conspicuous damage, nevertheless did not develop either. When part of the egg only is submerged, hatching often takes place, but with considerable delay.

Some preliminary experiments on the influence of lipolytic agents (ether, chloroform, benzol) in contact or as a gaseous medium, were begun too late (end of September) to permit of any conclusions. Apparently, in that late stage of development, phase *C* can be activated in this way. Yet proper experiments have still to be made.

Aqueous extracts of the material of the egg-pod wall, which some writers suggest. has an accelerating influence on development, did not show such effect.

SUMMARY AND CONCLUSIONS

Before giving the formal summary of our results, we should point out that two obscure points in the literature have been elucidated by this analysis: Uvarov⁷ (p. 229) in his discussion of the results of Plotnikov (1911) who obtained some larvae of *D. maroccanus* in autumn at temperatures ranging from 20–40° at high humidity, reports that development does not proceed at high temperature without moisture; it is obvious that "contact water" was meant by the term "moisture". Plotnikov suggested that under abnormally favourable conditions of temperature and rainfall larvae could hatch in

autumn. While, theoretically, that is not impossible, so far no report of such phenomena exists in which confusion with other species of grasshoppers is reliably excluded. That late autumn hatching can be produced quite freely in the laboratory is clear from our experiments. Yet the temperature as well as the rainfall regime of the typical areas of the Moroccan locust prevents the optimal autumnal combination from manifesting itself in nature. Bodenheimer¹ (p. 86 ff.) states that despite special scouting, he never observed autumnal hatching in Iraq. He points to another well-known phenomenon in that region: rains at the time of oviposition kill almost all the eggs. This phenomenon has been confirmed by our experiments in saturating freshly laid eggs with water; the experiments all ended with the early death of the eggs.

The normal egg development of *Doclostaurus maroccanus* was studied and described. Twenty more or less arbitrary stages were described, in which four main phases were distinguished:

1. Phase A, the initial period before formation of embryo;
2. Phase B, anatrepsis, which may continue for several months;
3. Phase C, readiness for katatrepsis; katatrepsis may be initiated when the eggs are wetted at temperatures above their developmental threshold;
4. Phase D, katatrepsis and completion of growth under favourable environmental conditions.

These four phases are evidence of an almost perfect adaptation to the climatic conditions of the natural habitat of *D. maroccanus*.

During the first three phases, retardation of development may occur which can be called "diapause". The preliminary experiments aimed at breaking the normal cycle, and the diapauses reveal that the normal trend of development is fairly strongly fixed by heredity and that those forces which are normally active in the natural environment of the Moroccan locust are unable to change the seasonal trend.

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ESTIMATION OF THE INCIDENCE AND DESTRUCTIVENESS OF PLANT DISEASES

I. TERMINOLOGY AND OUTLINE OF THE TECHNIQUE OF VISUAL APPRAISAL*

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A review of literature on plant disease estimation is concluded by proposing a number of terms to define the stages and objects of the process of estimation.

Visual appraisal of diseases is chiefly concerned with the degree, less with the type, of infection. Suitable criteria are the number of lesions or the area of affected tissue.

Disease effects such as tissue drying or leaf shedding cannot always be properly appraised.

The number of categories to be distinguished in appraisal, the choosing of proper units of appraisal and the problems of sampling are discussed. Outlines are given of scales of categories for plant disease estimation by small and larger units.

An outline of methods of evaluating data collected by appraisal and a discussion of the limitations of the use of estimations is to be published as the second part of this paper.

I. INTRODUCTION

The accurate determination of the effect of parasitic organism on their hosts is one of the primary concerns of phytopathology. In many cases such effect can be assessed without difficulty by measuring the yield of affected plants and comparing it with that of healthy plants, or by counting the number of affected plants. Frequently, however, the reaction of plants to disease attack cannot be measured in terms of definite numerical values, and the incidence of disease can be determined only by the estimation of leaf, stem or fruit area affected or by some similar expedient. Estimations of this kind have been applied to numerous phytopathological tasks, such as surveys, compara-

* Part II. of this paper will appear in a subsequent issue of this Bulletin and will contain the Bibliography of the whole article.

tive trials of varieties, ecological studies of pathogens, and fungicidal tests, and a great variety of methods has been worked out, especially in the case of cereal diseases. As the importance of estimations is generally recognized, it is surprising to find that there is very little critical literature on the subject. Methods of estimation applied to some particular cases have been described in a number of papers, but few attempts have been made, as far as we are aware, to coordinate such methods for general application. Notable exceptions are the scale of rust estimates used by the United States Office of Cereal Investigations, the instructions for the estimation of a considerable range of crop diseases published in Russia^{58, 66, 68, 74}, and the work of a committee of the British Mycological Society appointed to investigate methods of measuring plant diseases in the field.

The standarization of the procedure of estimations and of the methods of their evaluation, as demanded by Haenseler³¹, is a task of such magnitude that it should, in the writers' opinion, be reserved for an international convention of plant pathologists. The purpose of this paper is to state the problems involved in the estimation of the incidence and destructiveness of diseases, to point out some of the methods so far suggested for the purpose, and to discuss these in relation to the writers' experience gathered in field work in Palestine with many different crops. Unfortunately, the writers were somewhat limited in the range of American, and especially Russian, literature at their disposal, and the review of literature on estimations is, therefore, far from complete.

II. TERMINOLOGY

It is quite characteristic of the lack of attention paid to the subject of disease estimation that almost every author dealing with it has used different terms, or, even worse, has used similar terms with different meanings.

The term "estimation" is sometimes used to denote merely the recording of disease readings by visual appraisal, and sometimes in a wider sense to cover both these readings and the subsequent process of evaluation. Some writers have been concerned only with the technique of recording the incidence of disease in the field and of evaluating these records to determine the proportion of plant tissue affected by disease. For this purpose Tehon⁶⁹ and Tehon and Stout⁷⁰ distinguish between the prevalence of disease, defined as the percentage of stems or trees affected by disease, and what they call alternately the severity, intensity, or destructiveness of disease, i.e. the area of plant organs invaded by disease. Naoumov⁵⁴ similarly distinguishes between "*fréquence d'attaque*" and "*degré d'attaque*".

Other writers have emphasized that the taking of disease readings in the field is only one part of the process of estimation and that these readings have to be translated into terms of economic loss. Thus Brierley⁷ makes a distinction between the "extens-

ity" of disease, described as "largely a matter of distribution and rate of increase", and its "intensity" stated to be "largely a matter of lethality". Thus a disease prevalent in a crop without causing great losses may be regarded as of high extensity, but low intensity, whereas a disease causing heavy losses even when infection is restricted, possesses a high intensity. Ducomet and Foex¹⁶ distinguish between what they call "gravité apparente" of disease, which can be determined by taking readings of the percentage of diseased plant tissue, and "gravité réelle" or the physiological effect of disease on the plants. Similar and even more detailed distinctions are made by Naoumov⁵³. On the other hand, W. C. Moore⁵¹ and other British authors use the term "disease intensity" indiscriminately to describe the percentage of leaf surface, cereal heads or whole potato or beet plants affected by disease, without regard to the extent of losses incurred.

In order to facilitate the discussion of the problems dealt with in this paper, we propose the following terms:

Estimation is used in the wider sense of the word to cover the actual taking of readings to define the extent of infection, as well as the evaluation of the data collected in this way. The recording of the proportion of diseased tissue is termed *visual appraisal* or, briefly, *appraisal*. Visual appraisal is concerned with collecting data on the *incidence* of disease, which is identical with what Ducomet and Foex¹⁶ termed "gravité apparente". The incidence of diseases is measured by its prevalence and by the degree, and sometimes the type, of infection. The term *degree of infection* is used in the sense proposed by Naoumov⁵⁴ to denote the area of plant tissue invaded by disease or, at times, the number of lesions per unit area of tissue. The term *prevalence* is used in the sense proposed by Tehon⁶⁹ to express the relative percentage of diseased and disease-free plants or plant organs. But the absence of disease may logically be regarded as nothing but the lowest possible degree of infection. If the term prevalence is restricted to express the relation only between this degree on the one hand, and all the other possible degrees of infection on the other, a separate term is required to indicate the relative preponderance of each degree of infection; for this purpose we propose to speak of the *frequency of occurrence*, or simply frequency, of each degree of infection.

The second step in the estimation of disease is the *evaluation* of the data collected by visual appraisal. This evaluation may be limited to the mere computation of the general level of the incidence of disease from the mass of individual records of the prevalence of disease and of the frequency of the various degrees of infection. But in many cases evaluation of the field data aims at assessing the loss caused by disease. To arrive at an estimate of this loss, a number of factors additional to the incidence of disease must, however, be taken into account. These factors are discussed in detail in the final chapter and comprise the stage of growth at which the disease appears, the rate of disease development, and the vigour of the host plant. Moreover, with parasitic diseases,

the losses incurred owing to a given incidence of disease depend on the destructive energy of the pathogenic agent, which we shall term the *intensity* of disease, in the sense proposed by Brierley⁷ but distinct from the meaning which Tehon⁶⁹ and Moore⁵¹ attach to this term.

Between them, the incidence, stage of appearance, rate of development and intensity of disease and the vigour of the host plant determine the *destructiveness* of the disease which we define as its capacity to cause losses. Similar distinctions with regard to the factors determining the losses caused by disease have been made by Naoumov⁵⁴.

III. THE TECHNIQUE OF VISUAL APPRAISAL

The incidence of root or collar diseases or of systemic diseases, such as those caused by virus, can usually be determined in the field by simple counts of the plants affected or by estimation of the number of plants affected within a given radius¹. These troubles affect the plants as a whole, so that their prevalence is identical with their incidence, and estimates of the degree to which infection has progressed will in general be necessary only in detailed parasitological studies. In the case of diseases appearing on leaves, stems or fruits, on the other hand, determination of the prevalence of disease, i.e., of the percentage of diseased plants or plant organs, does not usually suffice, as it imparts no information as to the degree of infection, which may vary from traces to extreme severity. It is, therefore, to these diseases that estimations by visual appraisal are generally applied.

The first difficulty encountered in most estimations of qualitative characters is the choice of a suitable criterion of estimation. This clearly depends to a large extent on the purpose of the estimation. If it is desired to define the reaction of the host to disease, as is often the case in varietal tests, the various types and phases of disease development as well as the degrees of infection and their frequency of occurrence will have to be taken into account. In most other cases, however, estimation aims merely at assessing the level of incidence and the destructiveness of the disease, and then only the prevalence of disease and the frequency of occurrence of the various degrees of infection need be considered.

The actual process of appraisal is primarily concerned with determining the degree, and sometimes the type, of infection. This, as will be shown in the section on the categories and units of appraisal, is strictly true where individual leaves or stems are appraised separately. The frequency of each degree of infection is subsequently determined by counting the number of leaves, stems or fruits, in each category of infection. Where, however, larger units of appraisal are used, both the degrees of infection and their respective frequency enter into the definition of the categories used for appraisal, (cf. below on p. 89) and thus into the process of appraisal itself.

In order to elucidate the technique of visual appraisal, we shall first discuss the criteria which may be used in appraising the degree and type of infection, and shall then outline the use of these criteria in devising the categories of appraisal, the problems inherent in the choice of a suitable unit of appraisal and the problems involved in selecting truly representative samples for appraisal.

1. Appraisal by degree of infection of living tissue

The degree of infection may be appraised by the number of separate lesions on leaf, stem or fruit, or by the area of plant tissue invaded by disease.

A. NUMBER OF LESIONS

Appraisal by the number of lesions is permissible only in the case of diseases producing very small lesions of approximately uniform size; otherwise the variation in the size of lesions renders the data meaningless. Even where the spots are small, their number can be used as a criterion only if it is reasonably low. As long as this is the case, it is easier to state the degree of infection in terms of numbers of lesions than in terms of area affected. For this reason the lowest grade of infection in most scales of appraisal is conveniently defined by the number of lesions. In the writers' estimations of the incidence of rust (*Uromyces fabae*) of broad beans, for instance, the initial stage of infection was limited to "5 lesions per leaf", although the remaining categories of more severe infection were defined by the area of leaf affected⁶¹. Similarly a definition of "up to 2-3 lesions per leaf" was found to be suitable as a definition of the first category of infection by downy mildew (*Peronosplasmopara cubensis*) on cucumbers⁶⁰. Although the lesions produced by this fungus are larger, they are usually sharply delineated and rather uniform in size, so that the leaf area covered by 2-3 lesions does not vary excessively. In the case of other diseases, such as powdery mildew (*Erysiphe cichoracearum*) of cucumbers, the lesions grow large and indefinite. The number of these lesions may, therefore, be used as an indicator of the degree of infection only where a high level of accuracy is not essential, and even then only if a small number of lesions is involved.

Some authors, e.g. Roussakoff and Pantchenko⁶⁵ and Trumbower⁷³, have proposed scales for the appraisal of certain stem and leaf diseases in which the severity of disease is defined solely by the number of lesions. This procedure does not take into consideration differences in the size of leaves and would, therefore, appear to be less accurate than readings based on the proportion of leaf area affected by disease.

B. AREA OF AFFECTED TISSUE

In the appraisal of the incidence of diseases according to the area of affected tissue, the size of the lesions makes some difference in the procedure to be adopted. This has

been recognized by Tverskoy⁷⁴ who, in his instructions for the estimation of tobacco diseases, provides different scales of appraisal in accordance with the size of the lesion caused by each disease.

With all the diseases producing lesions of medium or large size, or small lesions readily coalescing into larger ones, only the area affected need be considered as a criterion of estimation. Where a high degree of accuracy is needed, this area is best determined in typical samples by the use of a planimeter, and the leaves (stems, fruits, etc.) to be appraised are then compared with these samples. This procedure has been adopted in all the cereal stem rust investigations of the Office of Cereal Crops and Diseases, U.S. Department of Agriculture, and in all the later work using the scale of estimations devised for these investigations. A similar procedure was followed by Tehon and Stout⁷⁰ in their estimations of a wide range of diseases of deciduous fruit trees.

In all these cases leaves or stems or parts of leaves or stems, affected to certain degrees considered typical of the various stages of disease development, were chosen as standards and were measured with regard to their total surface and to the area affected by disease. The resulting percentages of diseased area in such typical samples were adopted as categories in the appraisal of the incidence of these diseases. 4-6 such categories were so defined, the areas taken as criterion varying with each disease. Thus in Tehon and Stout's²⁰ estimations the categories for a disease like apple blotch (*Phyllosticta solitaria*), which produces smaller spots, ranged from 1.5% leaf area in the lowest, to 20% in the highest degree of infection; whereas the large lesions caused by the diffuse type of apple scab were estimated by categories ranging from 5% to 90% of affected leaf area.

In many cases, however, a rougher classification of affected leaves, stems or fruits will be found to suffice for estimation. Dillon-Weston¹³ in his appraisal of the incidence of yellow rust (*Puccinia glumarum*) of wheat distinguished merely between leaves free from rust, those affected on up to one quarter, on one to three quarters, or on more than three quarters of their surface. The Russian writers^{58, 60, 68, 74}, in their standardized instructions for the estimation of plant diseases, likewise distinguish only between 4 degrees of infection in general, defined as up to $\frac{1}{10}$, between $\frac{1}{10}$ and $\frac{1}{3}$, between $\frac{1}{3}$ and $\frac{2}{3}$, and more than $\frac{2}{3}$ of leaf or fruit surface affected. Similar scales have been found serviceable in our field work with a variety of vegetable diseases^{60, 61}, and in the estimation of the incidence of *Plasmopara viticola* on vine⁵⁹. Rough classification of this kind is also appropriate where the estimation aims at grading agricultural produce (fruits, tobacco, etc.) according to its market value. Here the only distinctions necessary are sometimes (a) traces of infection, which do not reduce the market value; (b) slight infection, reducing the produce to second or third grade; and (c) severe infection, which renders the produce unfit for marketing. Scales for the appraisal of

fruit diseases on the basis of affected area have been described by Marsh⁴⁵ in his review of methods used in Britain for the estimation of apple scab (*Venturia inaequalis*), by Blackford⁵ for citrus fruits affected with black spot (*Phoma citricarpa*) and melanose (*Diaporthe citri*), and by other authors. A similar scale was used by McKinney⁴⁷ to appraise the incidence of scab (*Actinomyces scabies*) on potato tubers.

That the evolution of methods of appraisal requires that due considerations be given to the laws governing human vision has been stressed by Horsfall and Barratt³³ and by Tehon and Stout⁷⁰. Discussing the manner in which the human eye distinguishes relative proportions of normally coloured and of discoloured tissue, the former authors state:

"According to the Weber-Fechner law, the human eye distinguishes according to the logarithm of the light intensity. Hence the grades (=categories of estimation) should be based on equal ability to distinguish, not on equal disease. Below 50 per cent, the eye sees the amount of diseased tissue. Above 50 per cent it sees the amount of disease-free tissue".

Tehon and Stout⁷⁰ have specifically dealt with the estimation of a disease producing *very small lesions*. Referring to apple blotch (*Phyllosticta solitaria*), they point out that in the case of the very small spots caused by this disease "the tendency in visual appraising is to emphasize the number, rather than the combined area of the spots". With a disease of this type, the number of lesions thus appears to be more easily appraised than their area. As mentioned above, however, this type of appraisal fails to take into account that a given number of lesions may represent a widely varying percentage of diseased tissue, depending on the size of the plant organ (stem, leaf or fruit) appraised. This difficulty may be overcome, if entire organs are to be appraised, either by varying the number of lesions permissible in a given category in accordance with the size of the organ (the organs may for this purpose be divided into two or more size classes) or by keeping the number of lesions per organ constant, but making it fit various categories, according to the size of the leaf. Alternatively, the incidence of disease in such cases may be gauged by appraising the number of lesions not on whole organs of variable size, but on a fixed segment of area per organ. Tehon and Stout⁷⁰ have chosen the first of these alternatives, adopting two standard sizes of apple leaves and using in their estimations of apple blotch two scales of categories, which were identical in the percentage of diseased leaf area permissible in each category, but differed in the number of lesions.

2. Appraisal by type of infection or stage of disease development

The type of infection must be duly considered wherever the disease to be appraised may manifest itself in any of a number of forms. This is frequently the case where the virulence of disease attack is examined under different sets of environmental conditions,

or where the pathogenicity of one parasitic organism is tested on numerous varieties of a given crop.

An example of a disease appearing in a number of different types, denoting different levels of virulence of attack, is cited by Rashevskaja and Vatsinin⁵⁸. These authors, dealing with the incidence of *Bacterium malvacearum* on cotton, distinguish between 4 types of infection according to whether the disease manifests itself as leaf spot, on the midrib of leaves, or on the leaf stalk, or whether it causes the leaf stalk to break. Similar distinctions may often be made in the estimation of *Phytophthora infestans* on potatoes, as this fungus may also appear in the form of leaf spots, or may affect the petioles or shoots. The difficulties arising in the evaluation of estimates of different types of infection will be dealt with in a later section.

The problem how to appraise varietal response to diseases has in the past received special attention in the case of certain cereal rust diseases. Stakman and Levine^{67a} have devised a scale of estimation to ascertain the reaction of wheat varieties to stem rust, and this has been accepted by various other authors^{20, 37}. Stakman and Levine distinguish between the types of infection met with in varieties of differing susceptibility, and the *degrees* to which these types have developed. Each type of infection is characterised by the size and frequency of rust pustules and by the reaction of the host tissue. "Moderate susceptibility", for instance, is defined as

uredinia medium in size; coalescence infrequent; development of rust somewhat sub-normal; true hypersensitiveness absent; chlorotic areas, however, may be present.

Four degrees (trace, slight, moderate, considerable), to which each type of infection may have developed, are then distinguished, "considerable" being defined as

infection better than normal; uredinia fairly numerous and scattered.

Main and Jackson⁴⁴, working with leaf rust of wheat, do not distinguish between type and degree of infection, but have devised a single scale of estimation in which both these components enter into the definition of each of the 5 classes of susceptibility; in this scale, "moderate susceptibility" is defined as

uredinia fairly abundant, of moderate size; no necrosis produced, but sometimes slight chlorosis immediately surrounding the uredinia.

Wei⁷⁵ in his work on the resistance of the garden bean to rust (*Uromyces appendiculatus*) distinguishes only between 4 types of infection, without considering the degree of their development.

Goulden, Newton and Brown²⁵, working with numerous races of *Puccinia graminis tritici*, have pointed out the difficulties encountered where the susceptibility of two varieties, varying widely in the type of their response to a given disease, is to be assessed by visual appraisal. Similar difficulties arise where several types of reaction occur in a single plant or variety. Several of the authors cited above^{67a, 75} therefore included a special type of "heterogeneous" infection in the scales of their estimations. There

seems to be no means of accurately comparing the susceptibility of plants exhibiting a fundamentally distinct or a heterogeneous type of response.

Scales of estimation devised according to the stages of development of a given disease may be applied to root and collar diseases. Thus, the extent of infection of cereals by the root-rot caused by *Helminthosporium sativum* has been estimated by McKinney⁴⁷ and by Greaney and Machacek²⁸ according to the degree to which the rot has progressed in various parts of the seedling, and Bremer et al.⁶ have similarly estimated the development of club-root of crucifers.

No generalisations can be made with regard to the categories to be used in estimations of various types of disease development or host response, as these depend entirely on the nature of the disease.

3. *Appraisal by proportion of dried tissues and of leaf shedding*

Most leaf and stem diseases, and some fruit diseases, ultimately effect the drying up of the affected tissues, and diseased leaves are often shed prematurely. This phenomenon introduces considerable difficulties into the technique of estimation. The causes of drying up or of leaf shedding are frequently not ascertainable after the event, at any rate not under field conditions. It is therefore self-evident that these last, and economically most important, stages in the development of many diseases can be estimated only if the drying up or leaf shedding is due solely to the disease in question and to no other contributory causes. Where only a small portion of tissue has dried up, the surrounding tissue often affords a clue to the causes of the drying. But if larger portions have dried up, no such clues are usually available and the amount of affected tissue cannot be satisfactorily included in any scale of estimation, unless the cause of the drying up is abundantly clear from other considerations. This may conveniently be illustrated by the example of the powdery mildew of tomatoes (*Leveillula taurica*), the incidence of which has been estimated by the writers on many occasions⁶². The bright yellow lesions caused by this fungus begin, after some time, to dry up from their centre outwards. There is no difficulty whatever in correctly appraising the incidence of this mildew as long as some part of the lesion still retains its yellow colour, but there is no means of diagnosing it accurately under field conditions once the entire lesion has dried up. Now individual lower leaves of tomato plants often dry up quite suddenly for reasons independent of leaf diseases, and these leaves are indistinguishable from those killed by mildew. Reliable data on the incidence of the latter could only be obtained if all the lower leaves were omitted in the estimation. On the leaves of the upper portion of tomato plants, on the other hand, drying up for causes other than leaf diseases is comparatively seldom met. In the absence of other diseases, therefore we, had no hesitation in estimating the incidence of powdery mildew on the upper portion of the

plants, even where some of the leaves had dried up completely and did not in themselves exhibit clear symptoms of the disease.

Similar difficulties have often been encountered in our cereal disease estimations. The lower leaves of cereal plants usually dry up some time after flowering; when attempting to estimate the incidence of *Septoria* leaf spot, we found it extremely difficult, therefore, to determine whether this disease or other factors were responsible for the drying up of leaves. The same applies where yellow leaf rust (*Puccinia glumarum*) has been arrested in its uredo stage by unfavourable climatic conditions which are apt to cause the drying up of leaves.

Further complications arise where two or more diseases may be involved in killing the tissue or bringing about the shedding of the leaves. Working with bananas, Simmonds⁶⁷ in his estimates allocated half a leaf to each of the possible causes of death, where the decision regarding the major cause of death was difficult. In our field work we have constantly come up against this problem in the case of cucumbers⁶⁸. Under Palestinian conditions the powdery mildew of cucumbers (*Erysiphe cichoracearum*) is so omnipresent that leaves affected only by the more restricted downy mildew disease (*Peronosplasmopara cubensis*) are practically never met. While powdery mildew usually induces the gradual and slow drying up of leaves, the downy mildew may kill them within a few days. Estimations of the incidence of downy mildew would thus lose most of their value unless the leaf-killing effect of the disease is duly taken into account. We therefore decided, in all those cases in which the rate of development and incidence of downy mildew greatly exceeded that of powdery mildew, to attribute all drying up to the former, and to neglect the effect of the latter disease. Although a certain error is introduced in this way the general results obtained undoubtedly reflect the relative incidence of the two mildews much more accurately than if all dried leaves had been omitted in the estimation. But under conditions where there was no very obvious difference in the rate of development of powdery and downy mildew, the drying effect could not be ascribed with certainty to either disease, and the dried leaves had to be excluded from estimation.

In fungicidal trials aiming at the combined control of several diseases the estimation of the percentage of dried leaf or stem area or of leaf shedding may, however, be of value (Horsfall and Henberger³⁴), although the relative importance of each of the factors concerned cannot be ascertained.

In the estimation of diseases causing premature leaf-fall of fruit trees, the extent to which the disease — as distinct from other factors — is responsible for leaf-fall may sometimes be ascertained by counts of the leaves accumulating on the ground below the tree. We employed this procedure in our work on the leaf spot of olives caused by *Cycloconium oleagineum*⁵⁷. Periodic counts of the leaves present on selected and labelled branches indicate the percentage of leaf-fall over a given period; examination of

the leaves on the ground reveals the proportion of leaves that may be assumed to have been shed owing to disease attack.

It must be admitted that this assumption is based on probability only, as there is no actual proof that an infected leaf has in every case been shed because of infection. Moreover, external factors, especially winds, may largely affect the extent to which infected leaves drop. These considerations may limit the value of examinations of the degree of infection of shed leaves, but in our view the results thus obtained will still be more accurate than results neglecting the extent of leaf drop altogether.

4. The categories and units of appraisal

A. NUMBER OF CATEGORIES

The decision as to the number of categories to be distinguished in appraising the incidence of diseases, and the definition of each of the categories in terms of area of tissue or percentage of plant organs affected, must be made with due regard to the purpose of the estimation.

In the estimation of disease on leaves, stems or roots, cases may occur where only rough distinctions between diseased and more or less healthy plants are necessary, and the number of categories need then be small. But in the majority of cases the estimates are meant to be as accurate as possible, and the decision regarding the number of categories to be adopted, then presents certain problems. The precision of estimates may increase obviously with the number of categories, but this is true only as long as reliable distinction between the latter can readily be accomplished in the time at the disposal of the investigator. In practice the number of categories used does not usually exceed six, or at most eight.

As mentioned above, it is often desired to determine the reduction in the quality of agricultural produce brought about by disease. Obviously the number of categories to be distinguished in appraising the incidence of disease will then have to correspond to the number of market grades. But in other cases more accurate information is sought with regard to the area of produce affected by disease, and the choice of a suitable number of categories is then subject to the same considerations as in the case of leaf and stem diseases. Marsh⁴⁶ in his review of the methods used in England for the estimation of apple scab on the fruit has cited examples of either type of scale; some authors take into account only the market grade of the fruit and distinguish accordingly only between 2-3 categories of infection, while others appraise the incidence of disease by the area of fruit affected and use 5 categories.

As will be seen below, the choice of the unit of appraisal constituted an important factor bearing on the number of categories suitably employed in estimation.

B. THE UNIT OF APPRAISAL

The incidence of disease on a given plot may evidently be determined in two ways: by appraising separately the state of infection of a number of plant organs (leaves, fruits, stems) and calculating the average incidence of disease from the mass of data, in which case the unit of appraisal is small; or by appraising directly the incidence of disease on whole branches, plants, groups of plants or plots, in which case the unit of appraisal is larger. The fundamental difference between estimations by small and by larger units lies in the fact that in the former the categories of appraisal have to take into account only the degree of infection (e.g., the area occupied by disease on a single leaf), while the categories used in the appraisal by larger units have to make allowance for the prevalence of disease (i.e., percentage of infected plant organs or plants) as well as for the degree of infection.

With certain diseases only one degree of infection can occur; i.e., if the disease has appeared at all it invariably attacks the whole, or almost the whole, plant or plant organ; e.g., the effects of systemic diseases, such as some of those caused by virus and certain root rots, are simultaneously apparent on all parts of the shoot. Further examples of diseases affecting an organ as a whole are the smuts and the ergot diseases of cereals. In all these cases the prevalence of disease constitutes, in itself, a sufficient measure of the incidence of disease, and the individual plant or plant organ presents itself as the obvious unit of what is strictly speaking no longer an appraisal but a count.

In the estimation of fruit diseases, too, no unit other than the individual fruit is usually employed, as the degree of its infection is easily appraised.

But in the estimation of leaf and stem diseases the unit of appraisal is not so obvious. Here the choice of the unit, whether individual leaves or stems, plants or groups of plants, or even entire plots, depends in the main on the growth habit of the host, the purpose of the estimation, the degree of accuracy desired, and the time at the disposal of the investigator.

The growth habit or training of plants has an important bearing on the choice of the unit of appraisal. Thus where cucumbers or tomatoes are staked individually, each plant may readily be estimated separately and serve as a unit of appraisal. But where these crops are left unstaked or are staked collectively (cordons), other units will have to be employed. Similarly, forage crops, sweet potatoes and other crops forming dense mats of growth cannot be estimated by individual plants or branches and the units to be used in appraisal are then either individual leaves or entire plots.

The choice of the unit of appraisal evidently determines to a large extent the accuracy of the estimation. There can be no doubt that strictly accurate estimations are possible only if the unit of appraisal is small, such as a single, or part of a single, plant organ. No other units have, therefore, received any consideration in the detailed and accurate schemes of estimation outlined by Documet and Foex¹⁵ and other authors. But,

in the writers' experience, occasions frequently arise in phytopathological field work, when the appraisal of the incidence of disease is desirable but either there is not enough time for the laborious appraisal by small units to be applied to large fields, or else a very high degree of accuracy is not essential for the purpose of the estimation. Such is often the case in fungicidal trials, where slight differences in the effect of the materials tested are of little account, and the number or area of plants to be estimated is usually large. A more rapid means of indicating the approximate incidence of disease on each plot may then be used to advantage, and one way of attaining this goal is by the use of larger units of appraisal, such as individual plants or groups of plants, parts of plots, or even entire plots. The use of such large units is recommended by the Russian writers^{58, 66, 68, 74} for the estimation of diseases on larger areas and by Townsend and Heuberger⁷² for fungicide experiments.

The choice of the unit of appraisal further depends on the purpose of the estimation. This point may well be illustrated by the case of covered smuts of wheat (*Tilletia tritici*), and barley (*Ustilago hordei*); if it is desired to determine the incidence of smuts with a view to gauging the extent of freedom from smut spores to be expected in the seed yield, the ear must serve as the unit of appraisal, because the amount of smut on the seeds will be roughly proportional to the number of affected ears. Entirely different considerations apply, however, if the appraisal aims at assessing the fungicidal efficacy of seed disinfectants in controlling smuts; with exclusively seed- or soil-borne diseases, such as these covered smuts, each case of infection is due to a single insufficiently disinfected seed grain, and even if the plant in question bears 4 infected ears, the fungicide can be considered to have failed only in its action on that single grain. The grain, or the entire plant growing from it — and not the ear — should, therefore, serve as a unit of appraisal in this instance.

In the following paragraphs we shall outline the technique of devising categories for appraisal by smaller and larger units, and some of the problems arising in the practical application of all these modes of estimation.

(1) *Small units of appraisal*

With leaf, fruit, or stem diseases the elementary unit of appraisal is, of course, the whole or a portion of each affected organ. With diseases affecting either surface of the leaf independently of the other, this elementary unit may be each surface of every leaf. As outlined above, each stem, leaf or fruit is best estimated according to the area affected. Each category of appraisal denotes a definite maximum proportion of diseased plant issue. In the case of leaf diseases, for instance, the scale of categories may be as follows :

- Category (a) — the leaf is free from disease *healthy*
- Category (b) — not more than 2-3 lesions per leaf or up to 1/10
of the leaf surface affected *trace*
- Category (c) — not more than 3/10 of the leaf surface affected *light infection*
- Category (d) — not more than 6/10 of the leaf surface affected *moderate infection*
- Category (e) — more than 6/10 of the leaf surface affected, but
the leaf does not yet dry up *severe infection*
- Category (f) — the leaf begins to dry up *very severe infection*

A scale of this type was found convenient in our estimations of the incidence of downy and powdery mildew of cucumbers (*Peronospora cubensis*, *Erysiphe cichoracearum*)⁶⁰. The exact definition of each category is, of course, largely dependent on the nature of the disease. Thus there are a number of diseases which cause the entire leaf to dry up even though only a comparatively small portion of the blade is seen to have been previously affected, and this and other peculiarities of diseases must be taken into account in devising suitable categories for their appraisal.

As pointed out below, the principal difficulty encountered in the appraisal of diseases by such small units is that of taking really representative samples. If this obstacle is satisfactorily overcome, appraisal by small units is by far the most accurate way of assessing the incidence of diseases of leaves, fruits and stems. To be reliable, however, the number of units so appraised must be fairly large and the process is extremely laborious.

(2) Large units of appraisal

As mentioned above, the appraisal of the incidence of disease by units such as branches, whole plants, or even larger units, makes it necessary to devise categories of appraisal which take into account the frequency of the various degrees of infection among the component parts of the branch or plant. Naoumov⁵⁴ has lucidly pointed out the interdependence of the severity of the degree of attack on individual plants or plant organs on the one hand, and the relative proportion of plants or organs affected, on the other hand. He wrote :

Qu'à mesure que diminue le degré de l'attaque des parties données de la plante, le nombre des parties atteintes baisse aussi; bien plus, le nombre des plantes envahies subit lui même un décroissement; d'où l'on déduit la règle que la fréquence de l'attaque des individus donnés dépend d'une certaine façon de la fréquence de l'attaque des organes et que cette dernière dépend également du degré de l'attaque.

In the terminology adopted in this paper, this means that the prevalence of disease

is in some way interdependent with the frequency of occurrence of the various degrees of infection. This rule may be made even more general and in the case of most leaf and stem diseases it is probably true to say that the relative frequency of occurrence of each degree of infection is interdependent with that of every other degree of infection. If, for example, the majority of leaves of a plant are slightly affected with disease, then we may expect a more or less definitive, but smaller, number of leaves either to be free from disease or to be moderately infected, while only few leaves of the same plant will be heavily infected. If the majority of leaves are heavily infected, a certain number will at the same time either have succumbed entirely to the disease or will show only moderate infection, while there will be only few slightly infected leaves and none of them is likely to be free from disease.

This rule is valid only where all parts of the branch or plant are equally susceptible to disease. It does not apply where the susceptibility of leaves increases or decreases with their age, as in that case a sudden change in environmental conditions may induce heavy infection on the youngest (or oldest) leaves while the remaining leaves, which have outgrown or not yet reached the phase of susceptibility, may be comparatively free from disease. The rule is evidently not strictly applicable to fungicidal trials, where the leaves are always unevenly protected from disease because of the unavoidable imperfection of the degree of coverage affected by the treatments and because some of the leaves form shortly after one application and remain unprotected until the succeeding application, while other leaves happen to be sprayed promptly after their formation.

If, however, due allowance is made for exceptions, the interdependence of the prevalence of the various degrees of infection may serve as the guiding principle in devising categories of appraisal for branches, plants or larger units. The actual range of these categories, i.e., the combining of a certain proportion of parts in one degree of infection with that of another portion in a second degree of infection, etc., to denote a definite degree of infection of the whole, is of course entirely arbitrary and the definition of these categories is usually governed by considerations of practical convenience. Each category should represent a definite proportion of tissue area. But the definition of each category cannot be rigidly fixed, because the range of degrees of infection found among the parts of a single plant depends largely on the rate of disease development.

In general, in composite categories for appraisal by larger units, the degree of infection of all the component parts will be the more uniform the quicker the rate of disease. Under these conditions, when $3/4$ of the leaves of a plant are moderately affected by disease, $1/8$ may still be affected slightly, and the remaining $1/8$ severely, while hardly any leaves will be free from disease.

Conversely, where disease development is slow, the component parts of a plant may

differ widely in the degree of their infection and this has to be reflected in the wide range of degrees of infections that should enter into the definition of the composite categories. We may then often find about half the leaves of the plant moderately affected, while varying proportions of leaves are affected severely, slightly, very slightly, or not at all. It may, therefore, be stated that the range of degrees of infection of component parts necessary to make up categories for appraisal by larger units is inversely related to the rapidity of disease development.

If it is desired to draw up scales of estimation to serve for the appraisal of any one disease under all conditions, a large measure of elasticity has thus to be preserved in the definition of composite categories.

(i) *Individual branches or plants*

In the following, an example is given of the way in which composite categories, which we shall designate A—F, may be devised for the appraisal of the incidence of diseases by units such as individual branches or plants. As stated above, each category aims at representing, as nearly as possible, a definite area of affected plant tissue, but alternative definitions are provided for each category, because a given proportion of diseased tissue may obviously be due to many leaves, being affected slightly or to a few being affected more severely.

In most cases the branches or plants to be appraised vary in the number of their component parts (leaves). We cannot, therefore, define the categories of appraisal by fixed numbers of leaves affected to various degrees, but can only do so by the relative proportion of leaves so affected. It is thus generally impossible to define a category as "up to 5 leaves affected up to one third of their surface", because in a plant with a total of 30 leaves infection of 5 leaves is still only a slight infection, while in the case of a plant possessing only 10 leaves, infection of 5 leaves may be ranked as severe. For this reason the composite categories to be used in appraisal by larger units usually have to state the *proportion* of tissue rather than the number of leaves affected, and may be defined as "up to one third of the number of leaves affected on not more than one third of their surface", or some similar definition. In this way a certain element of inaccuracy is, of course, introduced into the appraisal, as the observer has to estimate the proportion of the number of leaves affected as well as their degree of infection. In general, this cannot be avoided, but in some instances it is possible to limit the number of leaves to be appraised on each branch or plant, for example, to the 10 topmost leaves per branch. We may then define the categories of appraisal more accurately by the *number* of leaves in each degree of infection (categories (a)—(f), p.88), and may restrict each category to a more or less fixed maximum area of tissue affected. The categories for appraisal by units such as individual branches or plants, categories A—F, may, therefore, be described in either of the following ways :

| Category | Definition by <i>proportion</i> of component parts in each degree of infection | Definition by <i>number</i> of component parts in each degree of infection |
|---------------------------------|--|---|
| A—healthy | All leaves free from disease [category (a)] | All leaves free from disease [category (a)] |
| B—trace or very light infection | None of the leaves affected to a higher degree than that defined by category (b) or Not more than $\frac{1}{2}$ of the number of leaves in category (b), $\frac{1}{4}$ in category (c), and $\frac{1}{4}$ in category (a) | None of the leaves affected to a higher degree than that defined by category (b) or Not more than 7 leaves in category (b), 1 leaf in category (c) and 2 leaves in category (a) |
| C—light infection | Not more than $\frac{3}{4}$ of the number of leaves in category (c), few leaves in category (d), the remaining leaves in category (b) or Not more than $\frac{1}{2}$ of the number of leaves in category (c), $\frac{1}{4}$ in category (d), and $\frac{1}{4}$ in categories (a) and (b) | Not more than 8 leaves in category (c), 1 leaf in category (d), and 1 leaf in category (a) or Not more than 5 leaves in category (c), 2 leaves in category (d), and 3 leaves in category (b) |
| D—moderate infection | Not more than $\frac{3}{4}$ of the number of leaves in category (d), few leaves in category (e), the remaining leaves in category (c) or Not more than $\frac{1}{2}$ of the number of leaves in category (d), $\frac{1}{4}$ in category (e), and $\frac{1}{4}$ in categories (b) and (c) | Not more than 7 leaves in category (d), 1 leaf in category (e), and 2 leaves in category (c) or Not more than 5 leaves in category (d), 2 leaves in category (e), and 3 leaves in categories (b) and (c) |
| E—severe infection | The proportion of diseased tissue exceeds that defined by category D, but only few leaves begin to dry up or Not more than $\frac{1}{2}$ of the number of leaves in category (e), $\frac{1}{4}$ in category (f), and $\frac{1}{4}$ in categories (c) and (d) | The leaf area infected exceeds that defined by category D, but at most 1-2 leaves begin to dry up or Not more than 5 leaves in category (e), 2 leaves in category (f), and 3 leaves in categories (c) and (d) |
| F—very severe infection | The proportion of diseased and dried tissue exceeds that defined by category E | The leaf area infected and dried up exceeds that defined by category E. |

In this scheme the definitions of categories A—F by the number of component parts in categories (a)—(f) were devised to represent a definitive area of affected tissue. Bearing in mind that categories (a), (b), (c), (d) and (e) were given above the respective definitions of disease-free, up to 1/10, up to 3/10, up to 6/10, and more than 6/10 of

the leaf affected, categories A—F will be found to have been devised on similar lines. Thus category B conforms to a maximum affected leaf area not exceeding $10/10$ ($7 \times 1/10 + 1 \times 3/10$), category C is not to exceed $30/10$ ($5 \times 3/10 + 2 \times 6/10 + 3 \times 1/10$), category D comprises up to about $60/10$ affected leaf area ($7 \times 6/10 + 1 \times 10/10 + 2 \times 3/10$), and category E more than $60/10$ affected leaf area.

Another scale of estimation which adopts individual plants as units of appraisal has been proposed by Litwinov (as quoted by Naoumov⁵⁴) to determine the incidence of *Puccinia triticina* and *P. glumarum* on wheat. Taking advantage of the fact that these rusts develop first on the older, and then, progressively, on the younger organs of the plant, this author distinguishes between 9 categories of infection according to the number of the leaf on which symptoms have appeared (whether on the fourth, third, second, or first leaf from above) and according to the area occupied by disease on each leaf.

Individual plants have also served Horsfall and Heuberger³⁴ as units of appraisal in estimating the defoliation effect of *Alternaria solani* on tomatoes.

(ii) Groups of plants, parts of plots, or entire plots.

In many cases estimation by small units is impracticable, and the habit of growth, nature of the disease, or time at the investigator's disposal renders it equally difficult to use the individual branches or plants as units of appraisal. The corporate appraisal of groups of plants, parts of plots, or even entire plots, then affords the easiest means of assessing the incidence of plant diseases.

For this purpose if the categories described above for the appraisal of individual branches or plants (A—F) are defined in their more elastic form, according to the proportion of component leaves in categories (a)—(f), the scale may often be employed without change. If a given plot is then placed in category C, it is understood that up to one third of the total number of leaves of that plot falls into category (c), one third into category (d), and one third into category (b) cf. p. 89). This is the only means of appraising the incidence of diseases on crops where there is no means of isolating even individual branches for examination, e.g., in the case of tomatoes trained as cordons and forming a dense wall of growth. It has also been found satisfactory by Townsend and Heuberger⁷² for estimating the incidence of *Cercospora apii* on closely spaced celery.

Other crops, such as clover and lucerne, produce an abundance of slender stems, which may be adopted as units of appraisal. To obtain a representative estimate of the incidence of disease on plots of this type of crop, however, an extraordinarily large number of stems from all parts of the plot would have to be appraised. It is then more convenient to use larger units of appraisal, and the categories to be used may be based on the degree of infection, not of individual leaves, but of individual branches, stems,

or plants. These categories may be designated as A', B', C', D', E' and F' and they are defined in terms of categories A—F. In the appraisal by units such as groups of plants, parts of plots, or entire plots, the number of component parts (stems, plants) is hardly ever fixed, and the categories are, therefore, to be defined according to the proportion of parts in categories A—F. We thus arrive at something like the following definitions:

Category A' — healthy:

All stems (plants) free from disease (category A)

Category B' — trace or very light infection:

None of the stems affected to a higher degree than that defined by category B

or

Not more than $\frac{1}{2}$ of the number of stems in Category B, $\frac{1}{4}$ in category C, and $\frac{1}{4}$ in category A

Category C' — light infection:

Not more than $\frac{2}{3}$ of the number of stems in category C, isolated cases in category D, the remaining stems in category B

or

Not more than $\frac{1}{2}$ of the number of stems in category C, $\frac{1}{4}$ in category D, and $\frac{1}{4}$ in category A and B

Category D' — moderate infection:

Not more than $\frac{2}{3}$ of the number of stems in category D, isolated cases in category E, the remaining stems in category B

or

Not more than $\frac{1}{2}$ of the number of stems in category D, $\frac{1}{4}$ in category E, and $\frac{1}{4}$ in categories B and C.

Category E' — severe infection:

The proportion of diseased tissue exceeds that defined by category D', but only isolated stems begin to dry up.

Category F' — very severe infection:

The proportion of diseased tissue exceeds that defined by category D' and a considerable number of stems are drying up.

The writers wish to emphasize that the scales of appraisal set out above represent but an attempt to present the outline, couched in general terms, of a method of appraisal applicable to a great variety of crops and diseases. It must be clearly understood that far-reaching modifications of this method may be necessary in many cases, before satisfactory results can be obtained. Thus the nature of the disease and of the crop, and many other practical considerations, will have to decide the number of categories to be distinguished in appraisal, and the exact definition of each category in terms of leaf area affected or of the proportion of stems or plants affected.

The above method of defining composite categories for the appraisal of larger units, and even more, the use of these categories in the field, may at first sight appear very involved. Practice has, however, shown that observers can be trained within a relatively short time to employ this method of appraisal with a considerable degree of uniformity

and accuracy. The small units (leaves, etc.) may be compared to the letters of the alphabet, which combine to form words. Thus a beginner, both in reading and in estimation, may at first have to confine himself to the small units in order to arrive at the correct expression of their aggregate meaning. But once the eye has become accustomed to the process, even larger units may be appraised at a glance, much as we read a word without spelling out each of its letters.

5. SAMPLING

Sampling, in the estimation of diseases, is the choice of diseased plant material from a definite number of localities in a field, with the purpose of gauging the incidence of disease on the whole field according to the specimens chosen. The problem of sampling obviously does not arise where it is possible to appraise the whole plot directly, either by careful examination of each plant or by the more cursory examination which adopts the entire plot as the unit of appraisal. Correct sampling is of the greatest importance where individual leaves, stems or fruits, individual branches or plants, or groups of plants, are the unit of appraisal.

Taking samples involves three major problems concerning (1) the number of samples to be taken, (2) the size of each individual sample and (3) the mode of taking samples representative of larger areas.

It is evident that accurate estimation by sampling is possible only where the material to be sampled is reasonably uniform. The number of samples necessary for estimation depends primarily on the uniformity of the material and on the degrees of accuracy to be attained, being inversely proportional to the former and directly proportional to the latter.

The size of the sample, i.e., the number of separately appraised plants or plant organs included in a sample taken from one locality, depends largely on the prevalence of disease and on the unit of appraisal. Semiushina and Spangenberg⁶⁶ have pointed out that the size of sample must be larger, the lower the prevalence of disease, if the number of samples is kept fixed: if, for example, 20 samples are to be taken of a disease appearing on 1 plant in a thousand, we obviously have a better chance of obtaining a correct estimate if each sample consists of 100 plants instead of 10. Alternatively, the size of the samples may be kept constant but their number may be increased with much the same effect. The unit of appraisal has an important bearing on the size of samples to be taken at each locality. If the unit of appraisal is small (a leaf or a fruit), several such units may be combined in a single sample. If larger units of appraisal are used (individual branches, plants or groups of plants), each sample may comprise only one or at most two units when the disease to be appraised affects each part of the plant separately (e.g., leaf spots), or may consist of larger numbers of units, where the plant as a whole is affected (e.g., virus).

The principal difficulty encountered in sampling is that of ensuring that samples are (a) taken evenly from the entire area to be estimated, (b) chosen without bias and (c) include only comparable material.

Various methods have been proposed to make sure that the entire area under estimation is represented in the samples taken. In the field, samples are usually taken along a line traversing the plot under observation diagonally. If a higher degree of accuracy is desired samples may be taken along a number of lines traversing the plot either diagonally or lengthwise and crosswise. Moore⁵¹ has described the methods of sampling found most advantageous by British pathologists.

Sampling of diseased material is often extremely difficult, because the diseased plants or leaves tend to attract the eye rather more than do the healthy specimens. Samples are then often unreliable; and where the unit of appraisal is a branch, plant or group of plants, it may be preferable to take samples at fixed intervals throughout the field and not at random. Where the unit of appraisal is smaller, e.g., a leaf or fruits on the tree, no general rules can be stated as to the best method of sampling, because this depends on the growth habit and training of the crop. One case, in which the authors succeeded in solving this problem, is cited below.

It is of the greatest importance that samples be taken only from uniform material comparable in all respects. With many diseases the susceptibility of the leaf, fruit or plant varies considerably with age. Only samples of a uniform age composition can, therefore, be regarded as affording a genuine measure of the comparative incidence of such diseases.

The difficulty of taking samples both at random and from material of uniform age composition was encountered by the writers in their work with the downy mildew of cucumbers (*Peronoplasmodium cubensis*)⁶⁰. In Palestine cucumbers often develop luxuriant growth, but are not staked, and a fully grown row of cucumbers is a maze of intertwined vines. The downy mildew attacks first the older leaves and then progresses towards the distal end of the vine, to the younger ones. The purpose of our estimation was the determination of the relative efficacy of several sprays in controlling the diseases, and differences in the age composition of the samples estimated would have led to serious errors. We therefore adopted the following method of sampling: instead of picking separate leaves, entire cucumber vines were extracted from the maze of growth and each leaf on these vines was estimated separately. A definite number of vines along the whole length of each row was examined in this way. Owing to the fact that all the vines may be assumed to be alike in the age composition of their leaves, i.e., in the relative proportion of older and younger leaves, the uniformity of samples as far as age of leaf was concerned, was thus assured. In addition, the samples were truly taken at random, as the leaves were so intertwined that there was no way of telling which leaf belonged to which vine, until the vine had been extracted.

The foregoing paragraphs show that correct sampling is of fundamental importance for the accuracy of the estimation, as all the pains taken in laborious estimation by small units and in elaborate evaluation of the data cannot compensate for the error introduced by faulty methods of sampling.

IV. SUMMARY

- (1) The terminology so far proposed in literature on the estimation of plant diseases is reviewed and a number of terms defining the stages and objects of the process of estimation is proposed.
- (2) Visual appraisal, the term applied to denote the actual taking of records of the incidence of disease, is usually concerned with the degree of infection, less frequently with the type of infection or stage of disease development.
- (3) The degree of infection of living tissue may be appraised by the number of lesions or the area of affected tissue, the latter criterion being preferable.
- (4) Numerous diseases result in the ultimate drying up or premature shedding of the diseased organs. This effect can be estimated only if it is wholly, or almost wholly, due to the action of the disease; there is no way of assessing the incidence of disease accurately, if drying up or shedding is caused by more than one disease, or if other factors are partially responsible.
- (5) Although the precision of estimates may increase with the number of categories distinguished in appraisal, this is true only as long as reliable distinction between the categories is readily accomplished in the time at the investigator's disposal.
- (6) The choice of the unit of appraisal, whether individual plant organs, or part of such organs, individual branches or plants, groups of plants, parts of plots, or even entire plots, depends on the growth habit of the host, the purpose of the estimation, and the accuracy desired. Appraisal by small units is the most accurate, but is very laborious.
- (7) Outlines are given of scales of categories for the estimation of plant diseases by small and larger units.
- (8) The problems inherent in taking samples for appraisal are discussed. Correct sampling is considered to be one of the major factors affecting the reliability of disease estimations.

FRACTIONATION OF WHEY PROTEINS AS A MEANS OF GRADING MILK POWDERS AND DETECTING ADULTERATION OF FRESH MILK WITH MILK POWDER

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The differences between the various protein fractions of fresh milk and of milk made of milk powder are being investigated within the framework of an extensive research programme of "Products made from Fresh Milk and Reconstituted Milk and their Amelioration". On the strength of the data obtained so far, we have found it possible, even at this early stage, to indicate the way in which two urgent problems may be solved, namely, the grading of milk powders and the detection of adulteration of fresh milk with powder.

The usual method presently employed for this purpose is that of Evenson¹, with some modifications^{2,3}. The method is based on the principle that in the process of pulverisation of fresh milk into milk powder, carried out at comparatively high temperatures, the casein enters into combination with lactose (perhaps by condensation of amino groups), and possibly also with other decomposition products. This complex cannot be extracted from the casein precipitate by washing, and on addition of NaOH the yellow coloration, characteristic of sugars, is obtained.

Fairbanks² claims that as little as 5% of reconstituted milk*) in fresh milk may still be detected, but King³ concludes that 20—50% only may be detected with certainty, the proportion varying with the nature of the product, and recommends in his report⁴ that an effort be made to discover a more exact method.

The specifications of the American "Dry Milk Institute" for quality tests of milk powder include testing of acidity, solubility and centrifugal residue, but do not give a proper indication of the degree of denaturation of the powder, which is the decisive factor in appraising its quality for industrial purposes.

On the basis of the differences between the protein fractions of fresh and of reconstituted milk, found in the course of our work, we have developed a new method for the detection of adulterations and for the grading of milk powders, based on the fractionation of whey proteins.

* "Reconstituted Milk" is the product obtained by dissolving milk powder in water, in proportion of the total solids found in fresh milk, i.e. 1:9.

The Principle of the Methods

When milk is acidified by the addition of acid until the approximate isoelectric point of casein (pH 4.6) is reached, the casein, together with some other substances, is precipitated, while the albumins and the globulins (whey proteins), which will hereinafter be called "Fraction I", remain in the filtrate. As is well known, they can be precipitated from the filtrate by various means, e.g. by boiling. In milk which has been subjected to heat prior to precipitation of the casein, a part of Fraction I is carried down with the casein, and the quantity of the whey proteins obtainable from the filtrate on boiling decreases in accordance with the degree and duration of the heat treatment (see Rowland⁵). The quantity in mg% of whey proteins, which is precipitated on boiling Fraction I, will be referred to hereinafter as "Fraction II".

In view of the fact that, on being converted into milk powder, the milk is subjected to heat treatment by which its whey proteins are partially denatured, we have assumed — and our assumption has been confirmed by experiment — that the amount of Fraction II in the reconstituted milk will be much smaller than the quantity found on boiling the casein filtrate of fresh milk which has not been previously subjected to heat, [and it will often be practically non-existent]. We have, therefore, thought that the degree of denaturation of the powder — which, it must be assumed, is a very good indicator of the suitability of the powder for industrial use — may be determined by the fraction :

*Fraction II of reconstituted milk**Fraction II of fresh milk*

a formula which can thus be made to serve for the purpose of grading milk powders. On the other hand, if mixtures of fresh and reconstituted milk are prepared in varying proportions, it is to be expected that the amount of Fraction II precipitated on boiling the whey will be largest in fresh milk, and will decrease in linear (or almost linear) proportion to the decrease of the percentage of fresh milk in the mixture, and will be least — or even zero — in pure reconstituted milk. This is equivalent to saying that the reconstituted milk in the mixture serves, as it were, as a diluting agent, as if the fresh milk had been diluted with water; obviously, this may serve as a means of detecting adulteration of fresh milk with milk powder.

RESULTS**A. GRADING**

1. Reconstituted milk was prepared from samples of milk powders of the usual "high heat" type, made by different firms at different dates. Nitrogen in mg% was determined on the proteins precipitated from the casein filtrate on boiling — i.e. on Frac-

tion II —and the values thus found were much lower (0—4.5 mg% N_2) than the average values found for fresh milk (59.7 mg% N_2 ; see Table VII).

2. Samples of milk powders of the "low heat" type were examined. The heat treatment here is carried out at lower temperatures, and the values found were between 20—46 mg% N_2 .

3. All the above samples were examined by the method of Evenson and it was found that the result in respect to various powders of *the same* type (both in the "high" and the "low heat" type of powders) showed a considerable dispersion, as distinct from our fractionation method (see Table VII).

B. ADULTERATIONS

Mixtures were prepared of unpasteurised bulk milk, collected from various dairies on different dates, with milk powder of the usual "high heat" type, and the nitrogen content in mg% was determined on Fraction II.

The deviations from the average value of 59.7 mg% N_2 in pure fresh milk ranged from 3.5 to 4.9 mg% N_2 , i.e. 13%.

In respect to the mixtures, a continuous range of values was obtained between that for pure fresh milk and that for pure reconstituted milk. The deviations from the average values (4.3 to — 3.9) were almost equal to those found in fresh milk (see Table III).

We found that the official pasteurisation procedure (long pasteurisation: 63°C for 30 mins.; flash-pasteurisation: or the presence of preservatives [formalin, sublimate] 74—76°C for 15 secs.) does not affect the values obtained for unpasteurised fresh milk (see Tables V and VI).

All the above samples were also examined by the Evenson method. It was found that mixtures of the *same* sample of fresh milk with different samples of milk powder of the *same* type gave values altogether different from each other.

The reproducibility of the fractionation method is good.

EXPERIMENTAL

A. ADULTERATION

Reagents:

10% CH_3COOH ;
3% NaOH ;
 H_2SO_4 Conc + H_3PO_4 (3:1)
 CuSO_4 ; mixed with finely powdered Se
48% NaOH ;
4% H_3BO_3 ;
N/100 HCl

Indicator: 0.025% methylene blue in alcohol and 0.125% methyl red in alcohol (1:1)

PROCEDURE:

Precipitation of Casein

10 ml of the milk sample are placed in a centrifuge tube, heated on a water bath to 40–41°C and 3 ml 10% acetic acid added. The tube is left at this temperature for a further 10 min. to complete the precipitation. The solution is then centrifuged for 20 min. or filtered in the usual way.

Precipitation of Whey Proteins from Fraction I

10 ml of the whey protein filtrate, obtained by centrifuging or by filtration, is transferred to a small centrifuge tube and left on a boiling water bath for $\frac{1}{2}$ hour. It is then left overnight to complete the precipitation of the proteins and centrifuged on the following day. The precipitated proteins "I" are dissolved in a minimum amount of 3% NaOH (about 1–2 ml) and quantitatively transferred to a Kjeldahl flask.

Determination of Nitrogen in the Protein Fraction

To the protein solution in the Kjeldahl flask are added 3 ml of the $\text{H}_3\text{PO}_4 + \text{H}_2\text{SO}_4$ conc. (3:1) and, on the tip of the spatula a few dozen milligrams of $\text{CuSO}_4 + \text{Se}$ mixture. The mixture is then digested. When the digestion is completed, the solution assumes a greenish-blue tinge. 12 ml of NaOH 48% are added and the solution is steam-distilled into 5 ml of 4% solution of boric acid, containing mixed indicator (methylene blue-methyl red) in a Parnas-Wagner apparatus.

The results obtained are as follows:—

TABLE I.

Nitrogen (in mg%) Obtained from Fraction I on Boiling Mixtures of Fresh and Reconstituted Milk at 100°C for $\frac{1}{2}$ Hour.

| No. | Type of Powder Mixture | Fresh M. Reconst. M. | 100% 0% | 90% 10% | 80% 20% | 70% 30% | 50% 50% | 25% 75% | 0% 100% |
|-----|---------------------------|----------------------|---------|---------|---------|---------|---------|---------|---------|
| 1a* | 1-14-L Land-O-Lakes | | 66.0 | 51.0 | 46.0 | 43.0 | 24.0 | | |
| 1b | 1-14-L Land-O-Lakes | | 62.0 | 58.0 | 49.0 | 47.0 | 36.0 | | |
| 2a | Laurent Ide Dairy EST 471 | | 64.7 | 54.0 | 51.0 | | 28.5 | | |
| 2b | " " " " " | | 64.0 | 55.0 | 52.0 | | 29.8 | | 2.4 |
| 2c | " " " " " | | 59.1 | 50.8 | 38.3 | 38.1 | 35.1 | 19.5 | 3.0 |
| 3a | Eastern Star 470 | | 52.2 | 45.9 | 42.8 | 35.0 | 26.5 | 13.5 | 0.0 |
| 3b | " " " " | | 50.6 | 48.0 | 44.9 | 35.0 | 26.5 | 12.3 | 0.0 |
| 3c | " " " " | | 65.0 | 59.0 | 53.7 | 47.7 | 31.5 | 16.3 | 0.0 |
| 3d | " " " " | | 64.4 | 58.7 | 52.5 | 46.7 | 31.3 | 16.0 | 0.0 |
| 4a | Eastern Star 11.9 Haifa | | 56.2 | 50.7 | 46.2 | 41.0 | 27.8 | 16.6 | 4.8 |
| 4b | " " " " | | 55.3 | 50.4 | 44.5 | 40.3 | 27.6 | 16.6 | 4.6 |
| 4c | " " " " | | 63.2 | 57.1 | 51.8 | 47.4 | 33.5 | 18.7 | 4.3 |
| 4d | " " " " | | 62.1 | 56.3 | 50.6 | 47.4 | 32.8 | 18.4 | 4.3 |
| 5a | Nu Milk | | 61.1 | 52.2 | 47.8 | 35.8 | 25.8 | 15.7 | 0.0 |
| 5b | " " | | 57.8 | 51.8 | 46.4 | 35.5 | 24.9 | 15.4 | 0.0 |
| 5c | " " | | 60.0 | 54.8 | 50.0 | 41.6 | 28.3 | 13.8 | 0.0 |
| 5d | " " | | 60.5 | 54.8 | 49.9 | 40.4 | 28.3 | 13.5 | 0.0 |

* NOTE: Identical numerals marked with different letter indices indicate mixtures of the same powder with fresh milk from different sources. Similar results were obtained when mixtures

were prepared of corresponding proportions of fresh milk and water (in lieu of fresh milk and reconstituted milk).

TABLE II.

Fractionation of Mixtures of Milk and Water

| Mixture | Milk. Water | 100% 0% | 90% 10% | 80% 20% | 70% 30% | 50% 50% | 25% 75% |
|--|-------------|---------|---------|---------|---------|---------|----------|
| mg% N ₂ in albumin fraction Exp.1 | | 54.4 | 49.7 | 44.1 | 32.5 | 25.3 | not done |
| mg% N ₂ in albumin fraction Exp.2 | | 58.7 | 47.8 | 38.9 | 33.1 | 21.2 | 10.15 |
| mg% N ₂ in albumin fraction Exp.3 | | 58.6 | 48.5 | 40.0 | 33.0 | 22.5 | 10.0 |

TABLE III.

Summary of Average Values as Given by Table I. (Results Expressed in mg% N₂)

| Mixture | Milk, Reconst. Milk | 100% | 90% | 80% | 70% | 50% | 25% | 0% |
|---|---------------------------|------|------|------|------|------|------|------|
| | | 0% | 10% | 20% | 30% | 50% | 75% | 100% |
| mean Value | | 59.7 | 53.2 | 47.9 | 41.3 | 29.1 | 15.8 | 1.8 |
| mean Difference from Mixtures of Nearest Composition | | | 6.5 | 5.3 | 6.6 | 12.2 | 13.3 | 14.0 |
| mean Positive Deviation from mean Value | | 3.3 | 3.5 | 3.3 | 4.3 | 4.3 | 2.3 | 2.1 |
| mean Negative Deviation from mean Value | | -4.9 | -3.0 | -3.3 | -3.9 | -2.5 | -1.6 | -1.7 |
| Maximum Positive Deviation from mean Value | | | | | | | | |
| Maximum Negative Deviation from mean Value | | -9.1 | -7.2 | -9.6 | -6.3 | -5.0 | -3.5 | -1.8 |
| Standard mean Deviation | | 1.3 | 0.9 | 1.0 | 1.2 | 1.2 | 0.6 | 0.7 |

The results in Table I were obtained on boiling Fraction I for $\frac{1}{2}$ hour. Results obtained on boiling Fraction I for varying lengths of time are given in the following table:—

TABLE IV.

Influence of Duration of Boiling on the Degree of Precipitation of "I" (in mg% N₂)

| Mixture | Milk Reconstitd. Milk | 100% | 70% | 50% | 0% |
|------------------|-----------------------------|------|------|------|------|
| | | 0% | 30% | 50% | 100% |
| $\frac{1}{4}$ hr | Boiling | 58.5 | 39.1 | 28.8 | 0.0 |
| $\frac{1}{2}$ hr | Boiling | 60.3 | 41.5 | 32.1 | 13.2 |
| 1 hr | Boiling | 60.4 | 45.8 | 35.5 | 22.5 |

Preservatives. Samples of milk suspected of being adulterated were collected from various sources, mostly located at some distance from the laboratory. Preservative substances were then added to the milk, to prevent curdling. Table V shows the effect of these substances, which were added in their usual concentrations, on the precipitation of Fraction I.

TABLE V.

Influence of Added Preservatives on the Precipitation of "I" (in mg% N₂)

| Untreated Skim Milk | Skim Milk plus HgCl ₂ 10 mg% | Skim Milk plus 80 mg% formalin | Untreated Reconstituted Milk | Rec. Milk plus HgCl ₂ 10 mg% | Rec. Milk plus 80 mg% formalin |
|---------------------|---|--------------------------------|------------------------------|---|--------------------------------|
| 57.3 | 59.3 | 59.6 | 0.0 | 0.0 | 0.0 |

It will be seen that neither formalin nor sublimate in their usual concentrations has any appreciable effect on the amount of Fraction I. The influence of K₂Cr₂O₇ was not studied.

We also studied the effect of preheating fresh milk for various lengths of time and at various temperatures *under our experimental conditions*.

TABLE VI.

Effect of Preheating Fresh Milk at Various Temperatures and for Various Lengths of Time on the Degree of Precipitation of Fraction I.

| Temperature | — | 63° | 74° | 74° | 74° | 82° | 82° | 82° | 90° |
|--------------------------------|------|------|------|------|------|------|------|------|-----|
| Length of preheating | — | 30' | 5' | 10' | 15' | 5' | 10' | 15' | 5' |
| mg% N ₂ in fraction | 59.1 | 59.1 | 56.1 | 55.3 | 45.8 | 36.7 | 15.5 | 10.3 | 0.0 |

The curve corresponding to Table VI is similar to the well-known curve of Rowland (5). All determinations were carried out in triplicate.

B. Grading.

By Fractionation Method. Reagents: as above. Procedure: see above, except that for grading purposes 10 ml of whey are used for boiling (not 5 ml) on account of the small quantity of Fraction II.

By the Evenson Method, modified by us as follows: — after the washed casein precipitate is suspended in 5% NaOH, it is left for two hours and filtered through a Whatman 41 filter paper, and the colour intensity of the filtrate is determined in a photoelectric colorimeter, using a 420 m μ filter and a 5 ml cell. The colour intensity development by casein filtrate of fresh milk under these conditions is taken as G=100.

TABLE VIIa.

Determinations Made on High Heat Milk Powders.

| No. | Powder | Received on | Received from | % N ₂ in fraction "I" | "G", % light transm. (Even- son) | Remarks by dairies |
|-----|---------------------------|----------------|------------------|--|--|---|
| 1 | Laurent Ide Dairy EST 471 | 10.1.50 | Tnuva Rehovoth | 2.4 | 72.5 | Fair qual. for indu- strial purposes |
| 2 | Land-O-Lakes | 14.12.49 | Mikveh Israel | 3.5 | 58 | Average |
| 3 | Eastern Star 470 | 30.1.50 | Rehovoth | 0.0 | 48 | Below Average |
| 4 | Eastern Star 11.9.49 | 4.2.50 | Haifa | 4.5 | 79.5 | Fair |
| 5 | Yankee Made | 30.1.50 | Rehovoth | 0.0 | 80.3 | |
| 6 | Nu Milk | 4.2.50 | Haifa | 0.0 | 88 | |
| 7 | Cow & Gate | 6.2.50 | Dr. Gottfeld | 0.0 | | |
| 8 | East 416 carcier | 10.7.49 | Army | 0.0 | 73 | |
| 9 | Roller Australia | 6.2.50 | Dr. Gottfeld | 6.5 | 24.2 | |
| 10 | Unicef | 17.2.50 | Haifa | 0.0 | 58.5 | |
| 11 | Laurent 1st Grade | 14.6.49 | | 0.0 | 91 | |
| 12 | Laurent 1st Grade | 31.5.49 | | 0.0 | 92 | |

TABLE VIIb.

Determinations Made on Low Heat Milk Powders.

| No. | Powder | Received on | Received from | %N ₂ in fraction "I" | "G", % light trans. (Evenson) filter 420 mμ | Remarks |
|-----|-------------------------------------|----------------|------------------|---------------------------------------|--|-----------------------------------|
| 1 | Bacos, Australia | 17.2.50 | Haifa | 46.8 | 100 | Very good for ind. purposes |
| 2 | Cow & Gate | 1.3.50 | " | 20.1 | 71 | Good |
| 3 | Land-O-Lakes, low heated Dec. 49 | 29.3.50 | " | 43.9 | 100 | Good |
| 4 | Land-O-Lakes, plant 229, Jan. 50 | 29.3.50 | " | 42.6 | 100 | |
| 5 | Sanalac, Sanna Dairies | 18.4.50 | | 42.1 | 32.5 | |
| 6 | Land-O-Lakes, low heated | 17.4.50 | Rehovoth | 44.1 | 32.5 | Very good for ind. purposes |
| 7 | Land-O-Lakes, lot E 010, Feb. 50 | 10.5.50 | Paldorith | 31.7 | 67.5 | Good |
| 8 | Land-O-Lakes, lot E 0 5, Jan. 50 | " | " | 30.2 | 32.0 | |
| 9 | Land-O-Lakes, lot E 0 2, Feb. 50 | " | " | 32.7 | 52.5 | |
| 10 | Land-O-Lakes, lot 26, Jan. 50 | " | " | 37.1 | 18.8 | |
| 11 | Land-O-Lakes, lot 26, Jan. 50 | " | " | 39.0 | 44.0 | |

Interpretation of Results

A. ADULTERATIONS

It will be seen from Tables I and III that we were justified in assuming that by determining the percentage of nitrogen in the precipitate from Fraction I, the approximate proportion of milk powder added to fresh milk could be determined. The lowest proportion of milk powder which may be reliably detected by this method will depend on the deviations from the average shown by fresh milk. Considerable additional statistical material must yet be assembled for the purpose. In addition to this main factor (see Table III), the accuracy of the method will be established by rigorous standardisation of the procedure (the way of precipitating the casein and boiling the filtrate, the analytical method chosen for the determination of nitrogen, etc.). At the present stage of our work, 10–15% of added milk powder is the lower limit of detectable adulteration by milk powder of the usual "high heat" type. Where, however, at the time of examination, sample of fresh milk from the cowshed where the expected adulterator claims to have obtained his supply is made available to us for comparison, the effect of natural deviations is counterbalanced and the accuracy increased.

A number of factories have recently begun to produce a new, "low-heat" type of milk powder. In the samples examined by us the quantity of proteins in Fraction I had an intermediate value between that for fresh milk and that for "high heat" milk powder (see Table VII). The results obtained for mixtures show considerable deviations from the values obtained by the fractionation method. In such cases an appropriate procedure for the fractionation method must be found (perhaps lower fractionation temperature and shorter heating time, etc.). Preliminary experiments have shown, for instance, that with a heating time of 3 min. fresh milk gave a precipitate, but none was obtained from reconstituted "low heat" milk.

Comparison of the information on adulteration obtainable by our method with that obtainable by the Evenson method leads to the following conclusions:—

(a) At the present stage in the development of the fractionation method, neither method will detect adulteration with "low heat" milk powder.

(b) As regards "high heat" milk powder, the Evenson method will only detect adulteration qualitatively (see above), while the fractionation method will determine it quantitatively, within the limits mentioned above.

B. GRADING

The fractionation method gave values nearing zero in respect of "high heat" powders and 20–44 mg% N_2 in respect of "low heat" powders, while the Evenson method gave widely dispersed values (92–48) in respect of "high heat" powders and a similar dispersion (100–32) in respect of "low heat" powders. It would be premature to express

an opinion as to which of the two methods gives a truer indication of the quality of milk powder for industrial purposes. A number of appraisals were received from dairy workers (see remarks in Tables VIIa and b), but the appraising procedure should be standardised and its validity confirmed by industrial experiments.

SUMMARY

1. A method is described for the detection of the adulteration of fresh milk with milk powder, based on the determination of mg% nitrogen in the whey proteins precipitated on boiling the casein filtrate of milk.
2. As distinct from the Evenson method, now in general use, the method permits of a quantitative determination of the adulteration.
3. At its present stage the fractionation method will detect adulteration with "high heat" milk powders present in concentrations of from 10—15% upwards.
4. The principle of the method may be applied to the grading of milk powders of both types.

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INVESTIGATIONS ON d-LIMONENE, A BY-PRODUCT
OF THE CITRUS INDUSTRY

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The photo-oxidation of d-limonene was re-investigated at 110-120°. A ketone different from carvone has been isolated, suggesting the possibility of an oxidative attack in the 3 position of the d-limonene and opening the way for synthesis into the menthol series.

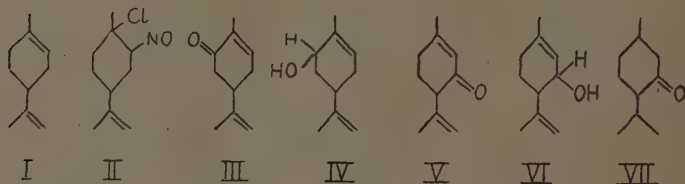
The increase in the manufacture of concentrated citrus juice — which in part takes the place of the marketing of fresh oranges — leads to the accumulation of certain by-products for which commercially feasible outlets have to be found; among these are the essential oils.

The essential oil of Israel oranges consists of more than 90% of d-limonene (I)¹, part of the balance (2%—5%) being composed of oxygenated substances. Owing to its unsaturated character, d-limonene is not stable to oxygen, moisture and heat, and orange oil deteriorates, quickly, therefore, upon storage. Its flavour becomes unpleasant and its practical value nil.

It has accordingly become an accepted practice to separate the more stable and valuable part of the oxygenated components of the orange oil by fractionation under diminished pressure or by distribution between two immiscible solvents². Thus, substantial quantities of d-limonene have become available, and a programme of research has been initiated with the object of finding uses for this unsaturated hydrocarbon.

The well-known uses of limonene, such as utilisation of its solvent properties³ or polymerisation or condensation to high-molecular products of the resin type⁴, hold out no promise, as the d-limonene of orange oil has no advantage, for these purposes, over the extremely cheap racemic compound dipentene, which is produced in large quantities from coniferae⁵. The present investigation, of which this paper reports the first results, is directed towards the production of optically active transformation products of high value, e.g., in the perfume, food or pharmaceutical industry.

Wallach⁶ has already observed that with d-limonene (I) nitrosyl chloride gives an addition product (II) which, by the classical procedure, can be converted into a ketone (III), l-carvone, which is the chief constituent of spearmint oil⁷. Even more important is the observation^{7a} that catalytic oxidation and also photochemical oxidation of d-limonene leads to the antipodic form, d-carvone (III), which is the active principle of the very valuable caraway oil⁸. Together with d-carvone, the corresponding secondary



alcohol d-carveol (IV) is formed. It is generally accepted⁹ that such autoxidation processes attack the "allylic" position in the immediate vicinity of the double bond, and we may, therefore, expect the isomer (V) of carvone and (VI) of carveol also to be formed in this process. The examples of Δ^3 -*p*-menthene¹⁰ and alkyl-cyclohexenes¹¹ have shown that methyl groups and tertiary carbon atoms are less prone to autoxidation than methylene groups.

Proctor and Kenyon¹² have pointed out that the infra-red absorption spectrum of fully oxidised orange oil is similar to, but not identical with, that of a mixture of carvone and carveol. Following this observation, which suggests the presence of oxidation products different from (III) and (IV), the photo-oxidation of d-limonene was re-investigated, and it was indeed established that ketones, other than carvone (III), are present in the product. At 110-120°, a ketone was formed which had the distinctive smell of menthone (VII) and gave a 2,4-dinitrophenyl-hydrazone of m.p. 202-203° (while those of d- and l-carvone melt at 192-193°). Equally, when the hydroperoxide, which is the primary product of this autoxidation process⁹, was reduced with sodium sulphite¹³ and subsequently catalytically hydrogenated, a secondary alcohol was formed which had the characteristic smell of menthol and boiled at 75-80°/4 mm.

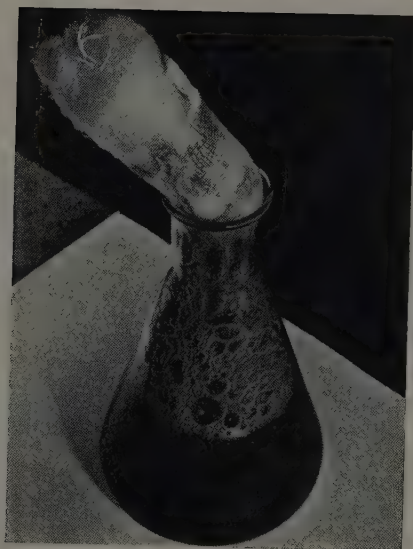
It is believed that these experiments have established the possibility of autoxidation of d-limonene in the 3-position and opened a new way into the menthol series. Menthol is an alcohol of great technical importance and a monopoly product reserved to few countries; its manufacture from d-limonene would constitute a valuable way of utilising this waste product of the citrus industry.

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Fig. 2. — (See page 116)



(See page 151)

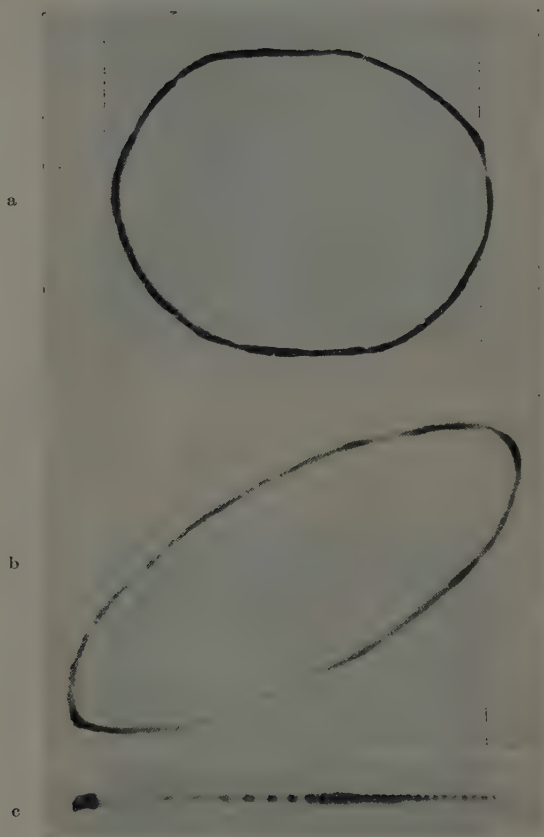


Fig. 3. — (See page 125)

LETTERS TO THE EDITOR

The Editorial Board does not accept responsibility for the views expressed in the letters printed below.

A PARADOX IN THE THEORY OF SAMPLING

Suppose that we have an urn with balls of which a fraction p are black and q white ($p+q=1$). A stranger who knows the general scheme but not the value of p is to be allowed to draw from the urn a sample of N balls, replacing each ball before drawing the next. If the sample contains m black balls the stranger will assert, using Bayes' theorem, that the probability that $|p-x| < \frac{1}{2}dx$ is $f_m(x)$ (say).

Let $\Phi_N(x,p) =$

$$\sum \left\{ f_m(x) \times \text{a priori probability that the sample contains } m \text{ black balls} \right\}$$

i.e. $\Phi_N(x,p)$ is the sampling expectation of the probability density that the stranger will attach to the proposition that p is in the neighbourhood of x . It may be shown that

$$\Phi_N(x,p) = (N+1)(x-q)^N P_N[(p-q)x+q](x-q)$$

where P_N is the N^{th} Legendre Polynomial.

If p is not equal to 0 or 1, $\Phi_N(x,p)$ has a unique maximum in the range $0 \leq x \leq 1$ at $x_{N,p}$, where

$$x_{N,p} = p + \frac{p-q}{2N} + \frac{p-q}{8pqN^2} + O\left(\frac{1}{N^3}\right).$$

Only for $p=0, \frac{1}{2}$, or 1 is $x_{N,p} = p$.

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A NOTE ON THE RESOLVING POWER OF THE FABRY-PEROT INTERFEROMETER

When a beam of light is reflected from the surface of a metallic mirror, it suffers a sudden change of phase. The change differs for light polarised with its magnetic vector parallel to the plane of incidence and light polarised perpendicularly to the plane of incidence. This differential phase change increases with the angle until it reaches a maximum value of π for grazing incidence¹. In the Fabry-Perot interferometer a phase change on reflection is equivalent to a change in the optical path distance between the plates. If such an interferometer is tilted to a sufficiently large angle relative to the direction of the illuminating beam, the differential phase change will cause each ring of the interference system to split into a pair of rings having mutually perpendicular states of polarisation.

In the normal use of the interferometer for spectroscopic work it is frequently necessary to tilt it to a small angle in order to bring a sufficient number of sharp rings onto the spectrograph slit. The angle is never large enough to cause actual doubling of the rings, but these may be slightly broadened. Tolansky has calculated that in certain extreme cases the broadening leads to a distinct reduction of the resolving power².

Although this reduction is very slight, we wish to report that we have actually observed it. We have used an interferometer having a plate separation of only

0.031 cm and it was found necessary to tilt it to an angle of 7 degrees with respect to the principal optical axis in order to obtain a suitable ring pattern. In a preliminary examination the instrument was illuminated by mercury green light and the rings were examined visually with very great care.

They were observed at infinity and also in the focal plane of a condensing lens having a focal length of 25 cm. No deterioration of the rings was observed as the interferometer was rotated from its position of normal incidence to that of 8 degrees. Sodium light was next used, the interferometer being set at an incidence of 8 degrees. The separation between successive orders was 15.89 cm^{-1} and the frequency difference between the sodium D lines is 17.18 cm^{-1} so that the D_1 and D_2 ring systems overlapped closely. They were actually indistinguishable but as the coefficient of reflection of the half-silvered plates of the interferometer was about 0.75 or slightly greater, they should have been just within the limit of resolution when observed visually. On the insertion of a polaroid disc behind the objective of the observing telescope, the doublet structure of the rings was plainly observed. It seems, then, that Tolansky's calculation is confirmed by observation and that it is indeed necessary to use polarised light in order to obtain the optimum performance from an interferometer set at non-normal incidence to the illuminating beam.

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COMPASS TELEMETERING

A method of telemetering a compass by radio has been developed. This system also makes possible utilization of the in-

formation for servo systems which are located at the sending or receiving end.

A narrow beam of light is reflected by a motor-driven rotating mirror, which is inclined at the required angle with respect to the axis of rotation (Fig. 1). The beam describes a cone on the face of the compass. Attached to the needle of the compass is a second plane mirror. Each time the rotating beam passes this mirror it is reflected into a photoelectric cell, which in turn produces an electric pulse. This pulse is amplified and after passing through an amplitude comparator is used to modulate a transmitter.

Simultaneously a *sine* voltage, originating from the motor system and synchronous with the rotation of the motor, is transmitted in order to provide the reference point for the display.

The motor system consists of a DC-motor and a three-phase generator on the same axis.

After separation of the *sine* and pulse voltages at the receiving end, a circular display is produced on the cathode ray tube by shifting the phase of the *sine* voltage by 90° . The original and the shifted *sine* voltages are applied to the X and Y plates, respectively, of the cathode ray tube, thus giving the circular display.

On this circular base two pulses are impressed during each cycle of the rotation of the motor: one originating from the photocell, the other—the reference pulse—derived from the received *sine* voltage by means of an amplitude comparator at the receiving end.

The reference pulse appears each time the phase of the *sine* wave passes through zero. (The negative pulse, resulting when the *sine* wave passes through π , is cancelled.)

These two pulses define an angle which gives the angular position of the compass needle with respect to a certain direction at the sending end.

The experiment shows that an easy reading is obtained with the reference pulse applied to the Y plates, and the

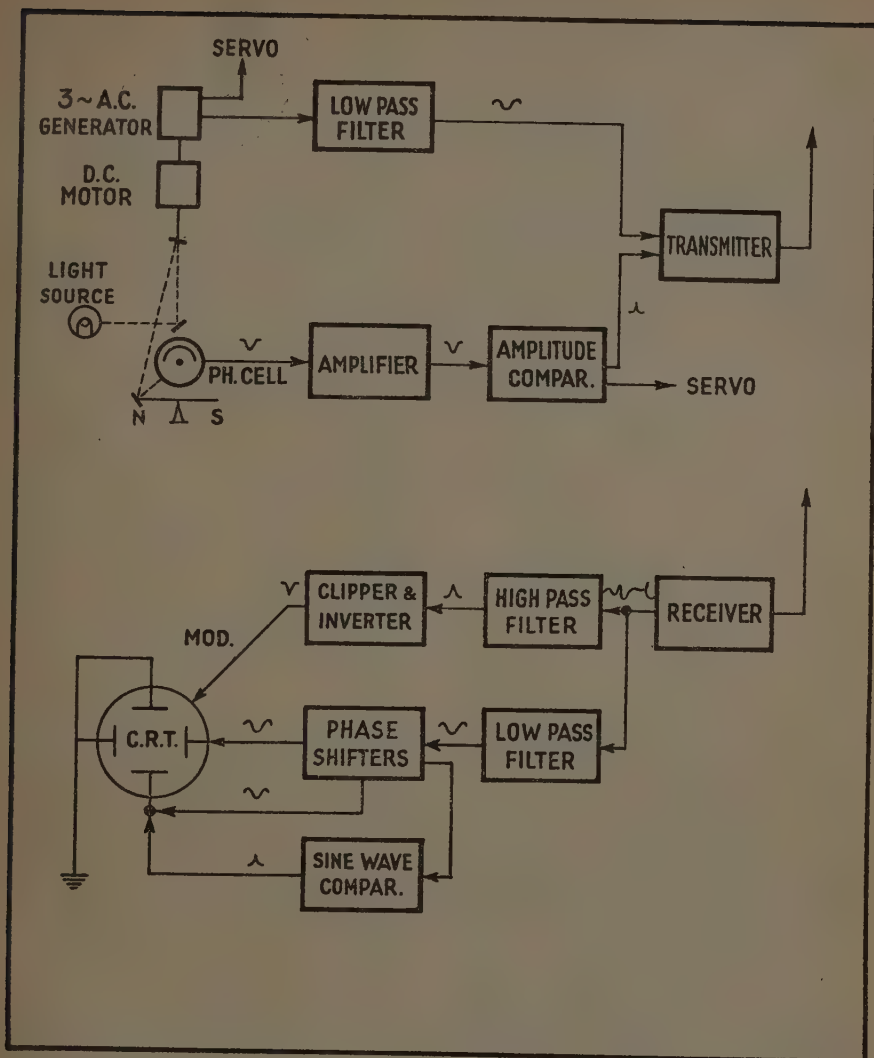


Fig. 1.

pulse defining the position of the compass needle to the intensity modulation electrode. The first gives a vertical line at the top of the circle, the second a dark spot at the angle defining the position of the compass (Fig. 2, see opp. page 113).

Another way of presentation is obtained by applying the reference pulse to the Y plates and by modulating both the anode voltage (sensitivity) and the grid bias (intensity) of the cathode ray tube with the pulse defining North. Thus a display with a vertical line at the top of the circle and a short radial line pointing inwards is obtained.

The error of the transmitted angle by this method is estimated to be $\pm 2^\circ$.

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EVIDENCE FOR THE NATURAL RADIOACTIVITY OF INDIUM

A consideration of the energetics of radioactive decay shows that two isotopes of the same mass number but neighbouring atomic number cannot both be stable, if the rest mass of the neutrino is zero. A number of such neighbouring isobaric pairs exist in nature; in some cases the expected radioactivity has been found in one member of the pair, with a period of decay comparable to or longer than the age of the earth (e.g. the well-known case of ^{40}K). The two naturally existing isotopes of Indium — ^{113}In and ^{115}In (abundances 4.5% and 94.5% respectively) are each members of such isobaric pairs, the first with ^{113}Cd , and the second with ^{115}Sn . Accordingly, it was decided to search for radioactivity among these elements, as none had hitherto been found.

Positive results were obtained with Indium. Using Geiger counters with easily changeable cathodes, a definite increase

in counting rate above the background rate was detected when Indium cathodes were used. The increase was of the order of 15 pulses per minute from a cathode of effective area 75 sq. cms and thickness 100 mgms./cm². Indium samples of different origin and of spectrographic purity were used and gave consistent results. The possibility that the counts were due to α particle contamination was excluded by the results obtained using proportional counters and from absorption measurements.

The author first tried to see whether the radiation from Indium could be identified with the characteristic K X-rays of Cadmium, which would be expected to follow the decay of ^{113}In to ^{113}Cd by K orbital electron capture. The experiments were carried out with krypton-filled counters which had an efficiency near to unity for these X-rays. The results were negative, and showed (in agreement with the results of previous workers¹) that the half-life for the decay of ^{113}In to ^{113}Cd by K-capture must be greater than 10^{14} years.

There remain two possibilities: — first, the decay of ^{115}In to ^{115}Sn by β -emission; secondly, the decay of ^{113}In to ^{113}Cd by L-orbital electron capture. Most of the radiation in fact was found to consist of β -rays. The half-value absorption thickness through aluminium under bad geometry was found to be about 10 mgms./cm², indicating that the energy of the β -rays must be of the order of a few hundred kilovolts. The interpretation of these results must be that ^{115}In decays by β -emission to ^{115}Sn with a half-life of the order of 10^{14} years.

The second possibility — that part of the radiation may be L X-rays following the decay of ^{113}In by L-capture — is of considerable interest. Outer electron capture should always accompany K-capture, but, in general, only to a small extent. It is consequently very difficult to detect, and in fact has only been observed in the case of A^{87} , using proportional coun-

ter technique². Under some exceptional circumstances, depending on the difference in energy and angular momentum between the two nuclei involved, it is possible that the probability of L-electron capture is greater than that of capture from the K shell. Using counters with different gas fillings and hence different efficiencies for cadmium L X-rays, results were obtained which were consistent with the hypothesis that a small part of the radiation consisted of cadmium L X-rays, but owing to the small counting rates involved the results cannot be regarded as conclusive evidence for this process. Assuming L-capture, one arrives at a half-life of the order of 10^{12} years for the decay of $^{48}\text{In}^{118}$ in this way.

In order to arrive at a more conclusive result on the L capture process, the use of Indium tri-methyl vapour as a filling in a proportional counter has been considered.

This work was carried out at the Cavendish Laboratory, Cambridge. I would like to express thanks to Prof. Frisch for his encouragement of this study. Further experiments (to complete this investigation), are under way at the Hebrew University, and it is hoped that a detailed account of the experiments will be published in the near future.

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THE EFFECT OF X-RAY INTENSITY ON THE RADIATION OF KMnO_4

The generation of ion pairs is considered to be the initial effect of X-rays, and the result of the irradiation of chemical com-

pounds is commonly expressed in numbers of affected molecules per ion pair generated. It is thus assumed that the X-ray effect is dependent only on the number of generated ion pairs, and the ion pair density (i.e. the number of ion pairs generated by the X-rays per unit of irradiated volume per unit of time) is not taken into consideration. The ion pair density, on the other hand, is proportional to the intensity, i.e. to the quantity of incident X-rays impinging on the unit of surface in the unit of time.

Some of our observations have led us to study the influence of the radiation intensity on the effect of X-rays. The substrate chosen for irradiation was a very dilute solution of KMnO_4 in dilute H_2SO_4 .

KMnO_4 in acid solution is known to be reduced to Mn^{++} salt under the influence of α or X-rays¹. The extent of this reduction (with sufficiently high doses of X-rays) can be easily estimated by titrating the KMnO_4 with $\text{Na}_2\text{S}_2\text{O}_3$ solution in the usual way before and immediately after irradiation.

According to some authors^{1,2} the quantity of KMnO_4 reduced is directly proportionate to the X-ray dose. Our results show that the extent of reduction depends not only on the total dose, but on the intensity of the irradiation as well. Thus, variation of the intensity of identical total doses of irradiation yielded varying results, and the reduction stands in an inverse relationship to the intensity of irradiation.

In our irradiation experiments, the cross-section of the X-ray bundle passing through the solution layer was always smaller than that of the layer, and the latter was thick enough for almost complete absorption of the incident X-rays. The absorption in the air between the anticathode and the solution surface being negligible, the intensity could be varied either 1) by varying the distance between the anticathode and the solution to be irradiated for application of equal doses in equal times; or 2) by altering the electron current through the X-ray tube: but in

this case irradiation time has to be varied accordingly in order to obtain equal doses.

Thus, on irradiating 50 cc of KMnO_4 approximately $n/250$ at two intensities varied with the distance, a dose of $3 \times 10^4 \text{r}$ reduced 1.3cc KMnO_4 $n/250$ at the intensity of $2 \times 10^5 \text{r/min.}$ and 2.0cc at the intensity of $1.68 \times 10^4 \text{r/min.}$

On varying the intensity according to method No. 2 a dose of $3 \times 10^4 \text{r}$ reduced 2.1cc KMnO_4 $n/250$ at the intensity of $1.56 \times 10^5 \text{r/min.}$ in 15 minutes, and 6.7cc at the intensity of $1.30 \times 10^4 \text{r/min.}$ in 180 minutes*.

As mentioned above, our results point to an inverse relationship between the intensity at which an X-ray dose is applied, and the extent of KMnO_4 reduced by it.

This phenomenon may be explained by assuming that, with increasing intensity of irradiation, there is a greater chance of the active radicals obtained combining with one another than with the solute molecules, since their number per unit volume is much increased, and this process would result in a diminished ionic yield.

KMnO_4 reduction by X-rays seems to be mainly an indirect reaction, at least under the conditions of our experiments.

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* The higher results obtained by varying the intensity with the current seem to be due to the difference in irradiation time. According to our experiments, KMnO_4 continues to be reduced even after irradiation is discontinued. Higher results should therefore be expected from prolonged irradiation than from short irradiation even if the doses are identical.

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DEPENDENCE OF THE ACTIVITY OF POTASSIUM CHLORIDE ON THE HISTORY OF THE SPECIMEN

During the thermal decomposition of potassium chlorate, a certain amount of perchlorate is formed^{1, 2, 3}. A study of the influence of the addition of potassium chloride and perchlorate on this thermolysis has been carried out. The amount of oxygen liberated was measured as a function of time, and the residue was analysed. The materials used were:

- I. Potassium chlorate (Baker's analyzed).
- II. Chlorate mixed with chloride which had been dried at 110° .
- III. Chlorate mixed with chloride which had been heated at 600° for an hour.
- IV. Chlorate mixed with chloride which had been obtained by decomposition of potassium perchlorate.
- V. Chlorate mixed with perchlorate and chloride which had been dried at 110° .

The following Table and Figure illustrate the substantial differences in the chemical activity of the various samples of potassium chloride at 453°C although X-ray analysis has revealed no differences between them.

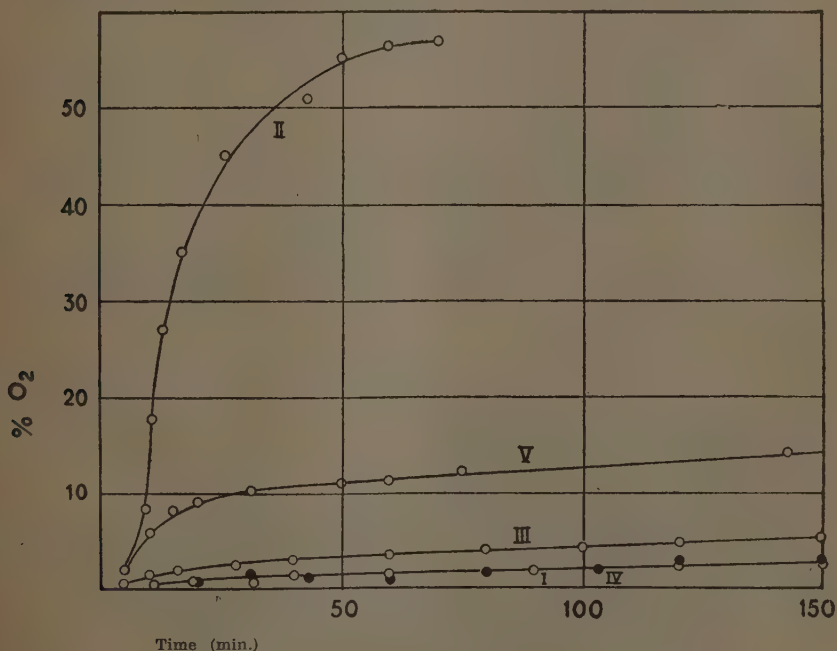
Potassium chloride (II), dried at 110° , accelerates the decomposition of chlorate, whilst samples (III) and (IV) have only a slight influence. Samples (III) and (IV), on the other hand, favour the formation of perchlorate (see the last column, KClO_4/O_2 , in the Table). Addition of perchlorate to the chloride dried at 110° (V) decreases the accelerating effect of the latter; this inhibition becomes more marked with increasing $\text{KClO}_4:\text{KCl}$ ratio.

Evidently the properties of potassium chloride vary with the history of the specimen. Thus, the chloride obtained from perchlorate (IV) or previously heated at 600° (III), absorb oxygen whilst untreated chloride (II) catalyses the decomposition of the chlorate according to:



Analysis of Decomposed Potassium Chlorate Mixtures
(All products expressed in mol-% chlorate).

| Sample | Initial Mixture | | Time of Reaction (min.) | O ₂ evolved | Residue | | | |
|--------|------------------------|-----------|-------------------------|------------------------|--------------------------|------------|--------------------------------|-----------------------------------|
| | Composition | Mol-Ratio | | | KClO ₄ formed | KCl formed | KClO ₃ undecomposed | KClO ₄ /O ₂ |
| I | KClO ₃ | 100 | 200 | 3.3 | 12.0 | 7.3 | 80.7 | 3.6 |
| II | KClO ₃ :KCl | 100:20.4 | 71 | 57.3 | 11.4 | 61.1 | 27.5 | 0.2 |
| III | KClO ₃ :KCl | 100:20.4 | 200 | 6.54 | 32.9 | 17.5 | 49.6 | 5.0 |
| IV | KClO ₃ :KCl | 100:20.4 | 200 | 3.55 | 33.85 | 14.85 | 51.3 | 9.4 |
| V | KClO ₃ :KCl | 100:25: | 228 | 14.8 | 37.3 | 27.2 | 35.5 | 2.5 |
| | :KClO ₄ | 37.5 | | | | | | |



Decomposition of Potassium Chlorate at 453°C.

It is believed that the inhibiting effect of the perchlorate on the chloride (V) is due to a deformation of the KCl lattice, thus enabling it to absorb oxygen and form perchlorate. A perchlorate-chloride mixture therefore behaves like KCl (III) and (IV).

The assumption that the potassium

chloride shows a "memory", is not without parallel. Weyl and Forland⁴ observed such effects in titanium dioxide crystals, for instance; and Hovi⁵ described a change in the lattice energy of mixtures of potassium halogenides when heated at 600°. Hedvall⁶ also suggested recently that a number of observations pointed to

the existence of "activated crystals", which show no apparent changes in the crystal lattice.

We are indebted to Dr. G. Schmidt (Weizmann Institute of Science, Rehovoth) for the study of the X-ray diffraction of our potassium chloride samples.

These experiments were carried out under the auspices of the Scientific Department, Ministry of Defence, and are published with its permission.

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THE SEPARATION OF ISOTOPES BY FRACTIONAL DISTILLATION

We have made a mathematical study of the process of isotope separation in a packed column. A dilute mixture of the required isotope is fed to one end of a packed column in which it is brought into intimate contact with vapour arising from a boiler situated at the other end of the column. Most of the liquid reaching the boiler is returned as vapour to the column, but a small fraction of the flow may be removed from the boiler as product. In

the column itself, there is an interphase transfer of isotope.

The isotope concentrations in the liquid, and the vapour at any point in the column, are functions of the distance from one end of the column and of the time. These functions satisfy certain partial differential equations which are easily written down along with the appropriate boundary conditions.

Solutions of these differential equations were obtained for the following cases:—

(a) finite supply reservoir operating without production; (b) infinite supply with production from the boiler. The solutions in the two cases lead to the following results:

$$q_B(t) = A_0 + \sum_{j=1}^{\infty} A_j e^{s_j t} \quad (1)$$

where $q_B(t)$ is the ratio of the concentration of the desired isotope in the boiler at time t to its initial concentration. The coefficients S_j are real and negative and so $A_0 = q_B(\infty)$ = the equilibrium enrichment =

$$\alpha e^{wZ} \left\{ R + B + Z \left(H + \frac{h}{\alpha} \right) \right\} \quad (2)$$

$$R + \alpha B e^{wZ} + \frac{H+h}{w} (e^{wZ} - 1)$$

where R and B are the capacities (in moles) of reservoir and boiler respectively, H and h are holdups (in moles/unit length) of liquid and vapour respectively, α = the unit process separation factor, Z = the height of the column and w is defined by the condition that αe^{wZ} is the equilibrium enrichment when working with an infinite supply reservoir.

The coefficients S_j are the roots of the transcendental equation

$$\frac{\tanh u}{u} = \frac{2}{wZ} \cdot \frac{\phi(1 + \theta s \tau) + \theta}{\phi \left\{ 1 - s \tau \left(\frac{1}{wZ} + \frac{\alpha + 1}{\alpha - 1} \theta \right) - \frac{\theta (s \tau)^2}{wZ} \right\} - \left[\theta \left(1 + \frac{s \tau}{wZ} \right) + \frac{2}{wZ} \right]} \quad (3)$$

where

$$u = \frac{1}{2} \sqrt{\omega^2 Z^2 + 2 \frac{\alpha+1}{\alpha-1} \omega Z s \tau + s^2 \tau^2} \quad (4)$$

$$\tau = \frac{Z(H+h)}{\text{Flow per unit time}} \quad (4a)$$

$$\phi = \frac{R}{Z(H+h)} \quad (4b)$$

$$\text{and } \theta = \frac{\alpha B}{Z(H+h)} \quad (4c)$$

An expression has been obtained for A_j ($j=1, 2, \dots$). In practice

$$|s_1| \ll |s_2| \ll \dots \quad (4d)$$

$$\text{and } |A_1| \gg |A_2| \gg \dots$$

and so the series converges very rapidly. Indeed for most purposes $q(t)$ may be adequately approximated by

$$q_B(\infty) - [q_B(\infty) - 1] e^{s_1 t} \quad (5)$$

(b) The general solution has the same form as before except that here

$$A_0 = \frac{\alpha(1+p)}{1+\alpha p} \cdot \frac{\frac{\alpha^2 p(1+p)}{\alpha-1} + 1}{\frac{\alpha^2 p(1+p)}{\alpha-1} + e^{-\omega z}} \quad (6)$$

where $p = \frac{\text{production}}{\text{rate of flow of vapour}}$ and w is the appropriate generalization of the definition given above (under a), and equation (3) becomes:

$$\frac{\tanh u}{u} = \frac{2}{\omega z} \cdot \frac{1 + \alpha p + \theta s \tau}{1 - s \tau \left[\frac{1 + \alpha p}{\omega z} + \frac{\alpha L + l}{\alpha L - l} \theta \right] - \frac{\theta (s \tau)^2}{\omega z}} \quad (7)$$

where u is defined by the appropriate generalization of (4) and L, l are rates of flow (in moles/unit time) of liquid and vapour respectively.

Equation (5) still holds.

Our results have enabled us to derive precise information about the values of α and ωZ from observations of $q_B(t)$.

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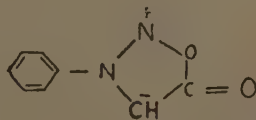
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THE CRYSTAL STRUCTURE OF THE SYDNONES. PART I.

This laboratory has begun an X-ray crystallographic investigation of the sydnones in order to obtain direct evidence on the molecular structure¹ of this class of compounds.



Through the courtesy of Professor W. Baker (University of Bristol, England) we have been able to examine crystals of N-phenylsydnone and of some of its derivatives, for which the X-ray data are recorded in Table I. From our data we may draw the following conclusions:

1. *Molecular weight*: The X-ray results are in good agreement with the chemical molecular weight. They do not, however, rule out a structure of twice the molecular weight and containing a centre of symmetry, since all derivatives investigated crystallise in centrosymmetric space groups.

2. *Molecular size*: From a comparison of the N-phenyl and the N-(p-tolyl) compounds, the volume occupied by one molecule can be estimated as $10 \times 5 \times 3.8 \text{ \AA}^3$, in agreement with the proposed chemical structure.

3. *Molecular shape*: The short 'b' axis in phenyl-sydnone and the equally short 'c' axis in the tolyl compound suggest that the molecules must lie near the (010) and the (001) planes, respectively, and that the benzene ring and the postulated five-membered ring system cannot be far from being co-planar.

Work on the structure analysis is now proceeding along two lines:

In an attempt to apply the Harker-Kasper-Gillis method^{2,3}, for determining the signs of the X-ray reflections, the intensities of tolyl sydnone were put on an absolute scale by comparison with those of oxalic acid dihydrate⁴. However, no

geometrical structure factors were found greater than 0.4. so that the inequality method could not be applied.

The similarity in packing of the phenyl and tolyl sydnones suggested the comparison of the Patterson projections $P(xz)$ and $P(xy)$ respectively in the hope of finding related vector patterns in the two projections. This analysis is now being undertaken.

The second line is the use of the heavy atom technique for structure analysis. The N-(p-chlorophenyl) sydnone being unsuitable, we prepared in this laboratory the m- and p-bromophenyl sydnones by the general method described by Eade and Earl⁵. Work is now proceeding on the three-dimensional analysis of the meta-compound.

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TABLE

List of X-Ray Crystallographic Data of Sydnones

| Compound | a | b | c | β | space group | spec. grav. | Z |
|---|-------|-------|------|-----------------|--------------|-------------|---|
| N-Phenyl-sydnone | 22.06 | 3.75 | 9.58 | $103^\circ 10'$ | $P2_1/a$ | 1.375 | 4 |
| N-(p-Tolyl) sydnone | 20.01 | 11.87 | 3.85 | $112^\circ 20'$ | $P2_1/a$ | 1.384 | 4 |
| N-(p-Chlorophenyl) sydnone | 16.9 | 13.95 | 9.37 | 129° | $P2_1/a$ | 1.531 | 8 |
| N-(m-Bromophenyl) sydnone | 12.37 | 9.13 | 7.45 | $97^\circ 30'$ | $P2_1/c$ | 1.9 | 4 |
| N, C-Diphenyl sydnone | 12.1 | 13.2 | 9.2 | 127° | $P2_1/a$ | 1.31 | 4 |
| N-(p-Chlorophenyl)- C-phenyl sydnone | 16.1 | 10.8 | 19.4 | 131° | Cc or $C2/c$ | 1.41 | 8 |

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TWO EXAMPLES OF THE USE OF THE FOURIER TRANSFORM METHOD IN THE ANALYSIS OF CRYSTAL STRUCTURES.

The Fourier Transform method^{1, 2} has been successfully applied to the determination of the molecular orientation of the naphthalene molecule in the unit cell³ and to the determination of the coordinates of a fixed point in the molecule of triphenylene^{4, 5}. We have extended the method to the determination of signs of structure factors in two cases, in one of which the position of the molecular centre is determined by space group symmetry, while in the second it had to be found with the aid of the transform.

In the crystal of phenazine ($a=13.22\pm 0.01$ Å; $b=5.061\pm 0.005$ Å; $c=7.088\pm 0.007$ Å; $\beta=109^\circ 13'\pm 7'$; $Z=2$; space group $P2_1/a$; confirming previous results⁶), the molecular centres must lie at the points $(0,0,0)$ and $(\frac{1}{2}, \frac{1}{2}, 0)$. The Fourier Transform of phenazine has been calculated assuming a planar molecule consisting of regular hexagons of side 1.40 Å. The structure is similar to that of naphthalene, and thus the method described in detail by Knott (loc. cit.) can be applied. For the $hk0$, $0kl$ and hkl spectra, it is of course necessary to take into account the fact that the two molecules in the unit cell are not parallel. In the $h0l$ zone the projected molecules are parallel, and hence the values of the structure factors at the various reciprocal lattice points may be read off directly from the transform. The signs obtained were used to calculate a 'b' axis projection, which was then refined in the usual way. The discrepancy, as commonly expressed, was 28% for F_0 obtained from the transform, while for F_0 from the final atomic coordinates it was 18%. Altogether 86 $h0l$ spectra were observed; for 78 of these the signs obtained from the transform agree with those calculated from the final atomic coordinates. This structure is now being refined. Until it is

completed no final assessment of the correctness of the other signs found from the transform can be made.

The cell dimensions of tetrabenzonaphthalene ($C_{26}H_{18}$) have been found to be as follows: $a=17.81\pm 0.02$ Å; $b=7.74\pm 0.02$ Å; $c=12.18\pm 0.01$ Å; $\beta=119^\circ 33'\pm 7'$ space group $P2_1/a$; $Z=4$. In the calculation of the transform of tetrabenzonaphthalene it has been assumed that the molecule is made up of regular hexagons whose sides are 1.40 Å, and that it is planar and centrosymmetric. As the centre of the molecule is not fixed by the space group, the expression for the $h0l$ structure factors takes the form

$$T_{obs} = 4 T_{h0l} \cos 2\pi(hu_0 + lw_0)$$

where T_{h0l} is the value read off the transform for reciprocal lattice points $h0l$ (as the molecules are parallel in this projection their contributions to the structure factor are all the same), and u_0 , w_0 are the fractional coordinates of the molecular centre.

The number of possible molecular orientations may be reduced to three by using the strong high angle spectra⁷; of these three possible orientations of the reciprocal lattice with respect to the transform axes, two may be eliminated because values read off the transform for certain $h00$ and $00l$ spectra are smaller than the observed structure factors. Thus the orientation of the molecule is approximately fixed. The coordinates of the centre are obtained from the $h00$ and $00l$ spectra in a manner similar to that described by Klug^{4, 5}. The values of the structure factors and their signs may then be found from the values read off the transform multiplied by the appropriate phase factors. These signs and the observed structure factors were used to obtain a projection down the 'b' axis, which is now being refined in the usual way. The approximation given by the transform in

this case is by no means as good as that obtained for phenazine.

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A MECHANICAL OSCILLOGRAPH FOR THE ANALYSIS OF THE ELASTIC PROPERTIES OF HIGH POLYMER SOLUTIONS.

The mechanical behaviour of high polymer solutions is often described by reference to a model composed of dashpots (viscosity), springs (elasticity) and mass (density). This allows a description of the behaviour of such solutions under small shearing (tangential) strains in the range in which the stress-strain function remains linear. The general model must consist of an infinite number of components. However, to describe the behaviour in a certain range of frequencies, four parameters are generally used^{1, 2} (see Fig. 1).

Various instruments have been built for the analysis of these parameters. For the range of 40-200 cycles, electric transducers have been employed^{3, 4} and the response of the electro-mechanical circuit measured electrically. For lower ranges an instrument has been built by Goldberg and Sandvik⁵ in which the liquid is contained between two concentric cylinders: the outer cylinder describes an oscillating movement, the inner one is held on a thin torsion wire; and the amplitude of the

oscillation of the inner cylinder is measured.

To measure the oscillation phase and amplitude (which gives more experimental data) and to define the range of

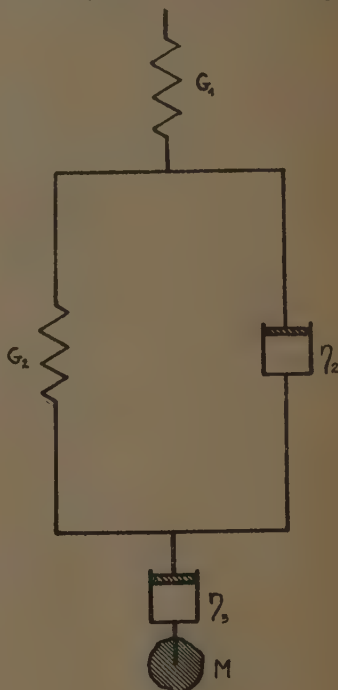


Fig. 1

linear behaviour of the liquids, a new instrument has been designed (Fig. 2). The liquid is again contained between two cylinders. The outer cylinder describes a harmonic oscillation, the inner cylinder is held on a precision miniature ball-bearing with very low friction. A mirror is mounted on each cylinder and a light beam from a concentrated-arc-lamp is reflected first by the mirror of the inner cylinder. The beam then passes through a rectangular prism in such a way as to turn the horizontal deflections of the first mirror into vertical deflections. The light is then

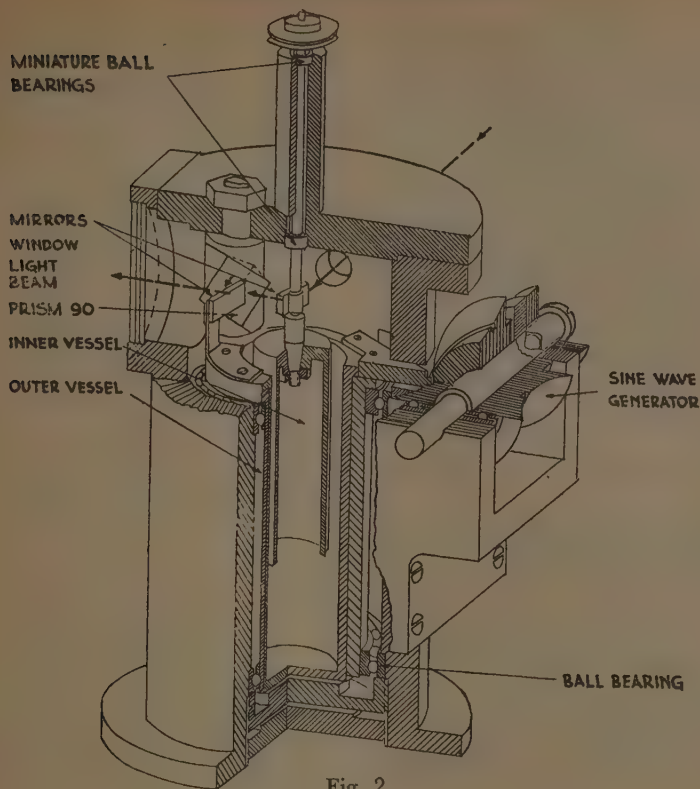


Fig. 2

reflected by the second mirror, and a lens system projects a picture of the light source on a phosphorescent screen of long persistence. If both mirrors describe harmonic movements of the same frequency, an ellipse results. Both amplitudes and phase differences between the cylinders (mirrors) can be measured in the whole range of frequencies (0.5-40 cycles). Non-linearity of the liquid will cause a distortion of the ellipse. By measuring the amplitude resonance frequency (Fig. 3a) and the frequency of the $\pi/2$ phase shift (Fig. 3b), parameters η_2 and G_2 can be calculated. It can be shown that

$$\eta_2 = G_2 \sqrt{\frac{1}{\omega_0^2} - \frac{1}{\omega^2}}$$

where ω = frequency of $\pi/2$ phase and ω_0 = resonance frequency of the undamped system. ω_0 can be calculated, according to Wazer and Goldberg⁶, from the resonance frequency and the ratio of the amplitudes at this frequency.

The moment of inertia of the inner cylinder and the distance between the outer and the inner cylinders can be changed by the use of different cylinders and loading disks. This permits a wider range of liquids to be tested, as well as the resonance frequency to be shifted into the desired range.

3% Latex solution in gasoline gave the following results:

$\eta_2 = 50$ poise; $G_2 = 4.7 \times 10^2$ dyne cm⁻²;

$\eta_3 = 73$ poise.

The same instrument can easily be adapted to the measurement of the viscosity and visco-elastic spectrum, thus serving as a constant stress viscosimeter. The outer cylinder is kept stationary, the inner is rotated by a weight giving a constant torque. After passing a chopping disc, mounted on a synchronous clock motor, the light is reflected from the mirror of the inner cylinder and projected to the screen or onto a photographic plate (Fig. 3 opp. page 112). According to the approximation method of Alfrey and Doty², the visco-elastic spectrum can be measured.

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CROSS-ELASTICITY

In 1948¹ I derived the most general law of elasticity in the form

$$\mathbf{p}_s^r = \mathbf{F}_0 \delta_s^r + \mathbf{F}_1 \mathbf{e}_s^r + \mathbf{F}_2 \mathbf{e}_s^r \mathbf{e}_s^r \text{ where } \mathbf{p}_s^r$$

is the tensor of stress and \mathbf{e}_s^r the tensor of strain. Many years ago, Poynting² found that a steel wire lengthens on excessive torsion and showed that a Hookean elastic solid, the equation of which is

$$\mathbf{p}_s^r = \mathbf{F}_0 \delta_s^r + \mathbf{F}_1 \mathbf{e}_s^r,$$

in finite simple shear exerts a pressure normal to the direction of the displacement and that the lengthening of the wire may be due to this property. Later Rivlin³ showed that a rubber cylinder, when

twisted between platens which prevent any change in length, exerts pressures upon them. The distribution of the pressure over the diameter of the circular end of the cylinder is not, however, in accordance with what is required by the Poynting effect. I have now shown, in a paper which will appear elsewhere, that the correct distribution can be found by taking account of the term containing \mathbf{F}_2 which I have named cross-elasticity.

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EXACT FORMULAS FOR PLATE AND CONE VISCOMETER

Piper and Scott¹ have described and made a rotating cone viscometer, but have assumed erroneously that the velocity gradient in the instrument is constant and equal to Ω/a , where Ω is the rotational velocity of the cone and a its angle with the plate. Recently Higginbotham² described a new plate-and-cone viscometer and emphasized that Piper and Scott's formula is correct only for very small angles α , e.g. $\alpha = \frac{1}{2}^\circ$. For the complete theory, see Mooney³. As it is difficult to use apparatus with an angle α so small that the approximation formula still applies, an instrument has been constructed in which $\alpha = 15^\circ$, and a new explicit formula has been derived for such larger angles.

The velocity gradient G varies with the angle Θ between the radius vector r and the vertical axis, but is independent of r , viz:

$$G = K / \sin^2 \Theta$$

where $K =$

$$\frac{\sin \alpha}{\cos^2 \alpha} + \ln \frac{\cos \frac{\alpha}{2} + \sin \frac{\alpha}{2}}{\cos \frac{\alpha}{2} - \sin \frac{\alpha}{2}}$$

The viscosity of the material, assumed to be Newtonian, is then

$$\eta = 3M / 4\pi R^3 K$$

where M is the torque and R the length of the side of the conical rotor.

This communication forms part of an investigation carried out under the auspices of the Scientific Department of the Ministry of Defence and is being published with its permission.

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PHYSICAL COMPONENTS OF A TENSOR

Murnaghan¹ appears to have been the first to point out that tensor analysis often fails to lead to quantities as measured experimentally. Such quantities have been named, not very aptly, the "physical components" of the tensor. Let V^i be a co-variant tensor component, then the physical component, according to Ollendorff², is,

$$V^i = V_i (g^{ii})^{\frac{1}{2}}$$

In certain hydrodynamical calculations in which Reiner's³ equation

$$p_j^i = F_0 \delta_j^i + F_1 e_j^i + F_2 e_\alpha^i e_j^\alpha$$

is involved, the physical components

p^{ij} , e^{ij} are required. Now it can be shown that if, in orthogonal coordinates, a tensor equation is written in mixed components, e.g. $p_j^i = f(e_e^k)$ and if the mixed components are converted into physical components, e.g. by

$$p^{ij} = p_j^i (g_{ii} g^{jj})^{\frac{1}{2}}$$

an equation of the same form, viz.

$$p^{ij} = f(e^{ke})$$

results. Therefore (always in orthogonal coordinates) these transformations are not necessary, and in our case we can simply write

$$P^{ij} = F_0 \delta^{ij} + F_1 e^{ij} + F_2 e^{ia} e^{aj}$$

making use of summation conventions for physical components and defining a "physical" Kronecker's delta by

$$\delta^{ij} = \delta_j^i = \delta_i^j$$

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THE EXTRUSION OF PLASTICS

Consideration of the equations of flow through an extrusion nozzle for a true fluid of viscosity η and a plastic (Bingham body) material of plastic viscosity η^* and a yield stress σ throws interesting light on the difficulty of forecasting the discharge from given geometrical variables and a single arbitrary technological property, such as "viscosity".

$$\frac{Q_{BR}}{Q_p} = \frac{\frac{\pi R^4 \Delta p}{8l\eta^*} \left[1 - \frac{4}{3} \left(\frac{2\sigma l}{R\Delta p} \right) + \frac{1}{3} \left(\frac{2\sigma l}{R\Delta p} \right)^4 \right]}{\pi R^4 \Delta p / 8l\eta}$$

$$= \frac{\eta}{\eta^*} \left[1 - \frac{4}{3} \left(\frac{2\sigma l}{R\Delta p} \right) + \frac{1}{3} \left(\frac{2\sigma l}{R\Delta p} \right)^4 \right]$$

where Q_{BR} is the quantity calculated in accordance with the Buckingham-Reiner equation and Q_p is that in accordance with Poiseuille's law and Δp is the pressure difference between the extremities of the nozzle, and R and l the half diameter and length of the nozzle, respectively. In the case where $\eta = \eta^*$

that is, in the case of closest possible similarity between the Newtonian and Bingham-bodies,

$$\frac{Q_{BR}}{Q_p} = 1 - \frac{4}{3} \left(\frac{2\sigma l}{R\Delta p} \right) + \frac{1}{3} \left(\frac{2\sigma l}{R\Delta p} \right)^4$$

It will be seen that if

$$\frac{1}{3} \left(\frac{2\sigma l}{R\Delta p} \right)^4 > \frac{4}{3} \left(\frac{2\sigma l}{R\Delta p} \right)$$

$$\text{or } 2\sigma l / R\Delta p > 4^{1/3}$$

$$\text{or } l/R > 2^{-1/3} \Delta p / \sigma$$

$$\text{then } \frac{Q_{BR}}{Q_p} > 1 \text{ and } \frac{Q_{BR}/\pi R^2}{Q_p/\pi R^2} > 1$$

The reverse holds true up to a certain limit.

In other words, by an arbitrary selection of the geometry of the extrusion nozzle in terms of the physical constants of a Bingham-body (which approximates to most plastics in the molten state) and of the applied pressure, the discharge and the average velocity of issue may be made larger or smaller than the corresponding discharge or velocity of issue of a true liquid.

This phenomenon, of which there has been no explanation to date, has been especially troublesome in connection with the extrusion of irregular sections, narrow sections at times discharging faster than heavier, contrary to all expectation.

A detailed account of this and related phenomena is appearing elsewhere.

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ON THE GENESIS OF CALCITE VEINS AND FLINT-BRECCIA IN THE UPPER SENONIAN OF PALESTINE

In the light of recent researches by the writer¹ which have proved the existence of erosional non-conformities between several stages of the Upper Cretaceous in Palestine, represented by relics of continental inter-stages, it seems possible to explain several hitherto obscure phenomena occurring in these formations. Here, the calcite veins and flint breccia occurring in the Upper Senonian are discussed.

The Senonian formation of Palestine, belonging mostly to its upper parts, is composed—as in most of the south-Mediterranean countries—of a lower chalky part assigned to the Campanian stage, and an upper part containing more or less extensive phosphatic layers and flint beds, assigned to the Maestrichtian stage. The

chalky limestone formation is of varying thickness, ranging from a few tenths to several hundred m. thickness (as in the Judean Desert). Characteristic of this chalk are the almost perpendicular, long, straight fractures, filled with calcite and appearing like veins which traverse the formation diagonally, mostly from SE to NW, and often also from SW to NW. The regularity of the veins leaves no doubt as to their connection with the tectonic development of the country. It is also remarkable that such veins are not visible in the strata below, but may continue irregularly in the phosphate beds lying above. It would appear that the agents responsible for the formation of the calcite veins have acted mainly upon the chalk formation.

In a recent publication² the writer has shown that the Senonian is framed below and above by erosional non-conformities, partly represented by small relics of continental formations. Thus, during Danian times most of the Senonian surface in Palestine was laid bare to atmospheric influences.

The beginning of the Senonian regression is manifested by the phosphate deposits and the flint-beds. The phosphate limestone is inherently a conglomeratic-detritic formation and was laid down in a very shallow sea penetrating into bays and between islands under very unstable conditions³. The frequent oscillations of this sea created a state of physico-chemical inequilibrium which from time to time provoked a sudden deposition of silica-gel, thus forming the extensive flint-beds.

It is obvious that the fractures in the Campanian chalk and their calcitic filling originated in the Maestrichtian but were completed during the Danian period. It is remarkable that in what Picard calls the "mottled zone" of the Danian the accumulation of calcite in veins and nodules is extremely rich, evidently owing to surface processes of those times. The emergence of the land in the Danian has

led to partial destruction and to fracturation of phosphate and flint beds and to the formation of the well-known flint-breccia. The agent of erosion and deposition in the Danian period was the existence of big inner basins which promoted deep penetration of calcitic solutions through the flint-breccia and the porous and shattered phosphatic beds into the fractured and partly denuded chalk formation. Thus were formed the regular calcite-veins of the chalk. The unconformed contact with the Turonian and the dividing continental deposits prevented deeper fracturation.

Further development on the Danian continent led to silica-cementation of the flint-breccia and often to advanced silicification of the phosphatic beds as well, exemplified especially by silicified phosphates of the Judean desert and of Transjordan. This explanation contributes to *completion* viz. modification of former theories⁴.

It may be further concluded that the tectonic events which influenced the formation of the Jordan Rift Valley are of ancient origin, dating from the end of the Cretaceous, but continuing almost up to the modern periods.

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LAWS OF MOTION OF WATER THROUGH SOILS

Darcy's law, governing seepage of water through soils, is extended to non-isotropic media. The permeability coefficient k becomes a 2nd degree tensor defined by 9 coefficients. In isotropic media the Laplace (potential) equation is valid, even for non-permanent flow. The boundary condition for the thoroughly moistened soil is constant (atmospheric pressure p_0). The seepage velocity at the boundary is then computed (from geometrical considerations) for the general, and some particular, cases.

Capillarity has the effect of reducing the constant pressure p_0 at the boundary by the amount of the capillary pressure

$$p = \gamma h \quad (1)$$

where γ is the unit weight of water, and h the capillary constant. One-dimensional horizontal flow obeys the formula

$$x = \sqrt{2 k h t / n}$$

where x is the distance travelled in time t by the liquid and n is the soil porosity. This equation is experimentally verified by visual and electrical means in a 4 m long horizontal glass capillarimeter, for both tap and Dead Sea water, and for sand and silt. The formula is valid initially in any direction. In this case one-dimensional vertical (up and down) and inclined flow were investigated. In vertical rise x tends asymptotically towards h (=capillary rise). In vertical descent the rate of seepage, initially proportional to $\sqrt{k h}$ tends asymptotically towards k (independent of h). If a static head H of water is available above the soil it should be added to h . In inclined flow the formula for vertical flow still holds, only t is to be multiplied by $\sin^2 \beta$ and x by $\sin \beta$ (β =inclination). Horizontal radial flow (two dimensional) was also studied.

When contact with the source of water

is interrupted there is no motion horizontally. Vertically, however, the moisture moves downward at a constant rate k/n .

Where the character of the soil changes in a horizontal direction the moisture moves from the soil of lower capillarity h' towards the soil of higher capillarity h'' . When $h'' \gg h'$, formula (2) may be applied (writing h'' , k'' , n'').

When highly capillary soil h'' overlies one of lesser capillarity h' , the moisture is suspended when its volume per unit horizontal area lies between $n''/(h''-h')$ and $n''/(h''-h')$, which are very narrow limits. Otherwise the moisture is drained.

The effect of air bubbles in the soil was also determined under isothermal conditions. When their pressure descends below $p_0 v_0/n$ (v_0 =initial volume of air in soil at p_0), the bubbles burst. This raises the capillary rise to $h/(1-v_0/n)$, and the hydrostatic pressure law is accordingly modified. Where the temperature increases with depth, the hydrostatic pressure has a maximum value.

On the above developments several experimental methods are developed: two for field permeability determinations in the undisturbed state (wide cylinder driven into soil and amount of water measured which maintains constant head; or rate of seepage measured by electrodes inserted into soil) and one for the measurement of air content in soil by means of piezometers.

Finally the density of moist non-swelling soils is computed, as well as their optimum moisture content. The advantage of expressing water content volumetrically is stressed.

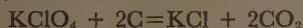
A more detailed account of this work, which is sponsored by the Research Council of Israel, is to be published elsewhere.

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CATALYSIS OF THE AIR OXIDATION OF CARBON BLACK.

In extending a previous investigation on the heterogeneous oxidation of carbon black by solid potassium perchlorate¹, the following observation was made: whilst at a total air pressure of 200 mm, only a very slow reaction occurs between carbon black and atmospheric oxygen at 367°C, almost all of the oxygen of the air is consumed in the oxidation of the carbon to carbon dioxide in the course of one to two hours, when potassium perchlorate is present. Concurrently with this *catalytic reaction*, normal oxidation:



takes place.

Potassium chloride has the same effect as potassium perchlorate, and this effect is independent of the previous history of the chloride².

A kinetic study of the reaction was carried out, using the apparatus previously described¹, except that the carbon dioxide formed was absorbed by ascarite in a cold limb of the reaction vessel; thus the conversion of oxygen to carbon dioxide could be measured manometrically. The samples were prepared from material ground to 200 mesh, mixed thoroughly, wetted, pressed into tablets under 600 atm. pressure and dried in vacuo, first at 120°, then at 250°. Small pieces of these tablets, containing 24 mg. of carbon each, were used for the experiments. The reproducibility of the results was satisfactory (deviation 2-3%).

Some of the results are given in the table below.

Oxidation of Carbon Black (24 mg carbon) by Air (p=200 mm, T=367°C) in the Presence of Catalysts

| Catalyst | Time (min.) | % O ₂ reacted | % C reacted |
|----------------------------|-------------|--------------------------|-------------|
| none | 120 | 7 | 0.8 |
| KClO ₄ , 2 mg. | 120 | 43 | 4.8 |
| KClO ₄ , 12 mg. | 120 | 68 | 7.7 |
| KCl, 2 mg. | 120 | 43 | 4.8 |
| KCl, 38 mg. | 120 | 63 | 7.2 |

Plotting $\log (a-x)/(b-x)$ versus t , [where $(a-x)$ =amount of carbon, $(b-x)$ =amount of oxygen present at time t], straight lines were obtained until the reaction was 65% complete. Thus the reaction appears to be of the second order, conforming to the equation $dx/dt=k(a-x)/(b-x)$. As in all cases of heterogeneous reactions, this equation has only limited significance, the value of k being dependent on the absolute quantities of the reactants.

This work is being carried out under the auspices of the Scientific Department, Ministry of Defence, and is published with its permission.

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THE CONFIGURATION OF PHENYLSELINE AND A NEW SYNTHESIS OF CHLOROMYCETIN.

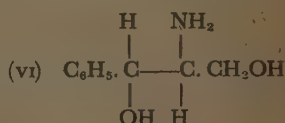
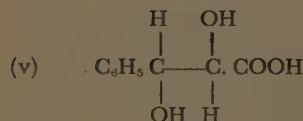
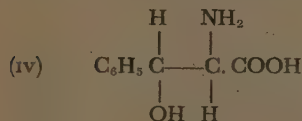
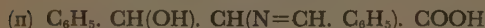
The constitution of the phenylserine (I), m.p. 190°C, obtained in the form of a benzylidene-derivative (Ia) from benzaldehyde and glycine in presence of aqueous alkali^{1,2} or of sodim metal in ether³ has frequently been questioned. The reason for this is that in the original synthesis an isomer (m.p. 230-232°C) was observed occasionally, which the discoverer, Erlenmeyer, considered to be an *iso*-phenylserine (II), while Foster and Rao⁴ assumed it to be the diastereomeride of (I) (two asymmetric carbon atoms!). Their evidence, however, was faulty: they obtain-

ed the same substance, *e.g.* from α -chloro- β -hydroxy-hydrocinnamic acid (III) with ammonia, a reaction which is not unequivocal, as the *direct* substitution of the chloro-atom has not been proved; indeed, it is most unlikely to occur⁵.

In can be assumed that the *constitution* of (I) follows from the synthesis, especially in its recently discovered⁶ modification in which no condensation agent is used and, therefore, the probability of unexpected reactions is minimized. However, the *configuration* of phenylserine (I) has not been established. The fact that (I) gives oxazoline derivatives with benzoic and acetic anhydride, respectively^{1, 2}, can be taken as an argument in favour of the *threo*-configuration (IV)^{7, 8}. The same conclusion has been reached by Billet⁹ who converted (I) into the known *threo*-phenylglyceric acid

(V); however, this method is open to criticism^{10, 11}.

It has now been found that the ethyl ester of (I), upon reduction with lithium aluminium hydride in tetrahydrofuran as solvent, gives *threo*-3-phenyl-2-amino-propane-1, 3-diol (VI) in acceptable yield (m.p. of the diacetate 165°C. — Anal. Calc. for $C_{13}H_{17}O_4N$: N, 5.6; Found: N, 5.6; 5.7). The literature data for the *threo*- and *erythro*-diacetate are 168-169° and 110-111°C, respectively¹². This reduction cannot cause an inversion of the configuration at the 3-carbon atom which is not involved at all in the reaction. *threo*-3-Phenyl-2-amino-propane-1, 3-diol (VI) can be nitrated according to Controulis, Rebstock and Crooks³, to give *chloromycetin*. The above observations constitute, therefore, a new synthesis of this antibiotic.*



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* Since this communication has been submitted, the reduction of the ethyl ester of (I) has been described by Carrara and Weitnauer (Chem. Abstr., 1950, 44, 7268) and by Vogler (Helv. Chim. Acta, 1950, 33, 2111).

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THE ISOTOPIC CHANGES OCCURRING ON THE FRACTIONATION OF METHANOL

The relative volatility of liquids of similar constitution depends essentially upon the magnitudes of the interactions between their molecules. These interactions, in the case of the non-polar liquids, are due to van der Waals forces, which depend upon the size of the molecule, its structure and polarisability but do not involve directly the masses of the atoms in the molecule. If we compare non-polar molecules differing only in isotopic composition, we shall not expect to find, to a first approximation, any difference in their volatilities (i.e. $\alpha=1$). It is only when second order effects are considered that small differences in the intermolecular interactions are predicted. The effects are very small except in the smallest molecules and at the lowest temperatures.

In polar liquids, however, particularly in those containing hydroxyl groups, additional relatively powerful intermolecular forces operate, namely the interaction of permanent dipoles. These are the forces usually associated with the hydrogen bond. Isotopic replacement of the atoms taking part in the formation of the hydrogen bond (most commonly H and O) will affect the strength of interaction to some extent and thus lead to a value of relative volatility of the two components differing from unity. It has already been suggested that it is these effects which are responsible for the possibility of separating the heavy isotopes of hydrogen in hydroxylic liquids by fractionation.

Isotopic substitution in parts of the molecule not participating directly in hydrogen bond formation will not be expected to affect the strength of the association and therefore will not give rise to components with differing volatilities.

To test this hypothesis further, we have subjected methanol to fractional distillation in a column which had already been proved suitable for isotopic enrichment in

the case of water. When an approximate equilibrium had been reached, samples from the top and bottom of the column were analysed for the isotopic content of carbon, methyl hydrogen, oxygen and hydroxyl hydrogen. It was found that no change in isotopic constitution of carbon or hydrogen of the methyl group had occurred, while the oxygen and hydrogen of the hydroxyl group were both enriched in the liquid phase with respect to their heavy isotopes, oxygen by a factor of 2, hydrogen by a factor of 16. These results are in agreement with those of Urey and co-workers¹, who report no C¹³ concentration, but found a two-fold increase in O¹⁸ on fractionation of methanol in the Pegasus column. Dedusenko and Brodski², obtained enrichment of O¹⁸ on fractionation of ethanol¹.

In the case of methanol, the following components must be considered: CH₃OH, C¹³H₃OH, CH₂DOH, CH₃O¹⁸H and CH₃OD, ignoring less probable combinations. The results show that isotopic substitution in the methyl group does not lead to separable components, the volatility ratio for the pairs CH₃OH:C¹³H₃OH and CH₃OH:CH₂DOH being apparently equal to unity, whereas for the pairs CH₃OH:CH₃OD; CH₃OH:CH₃O¹⁸H and CH₃OD:CH₃O¹⁸H the relative volatility differs from one. As in the case of water, we find here again that the effect of deuterium substitution is greater than that of O¹⁸.

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FACTORS GOVERNING THE RATE OF CRYSTALLISATION OF SUPER-COOLED MOLTEN TNT

It is known that the rate at which the crystals of an organic substance grow in its super-cooled melt is influenced by impurities; it has not been known, however, whether this rate-depressing influence depends only on the quantity of the impurities present or whether constitutional factors play a part. It would be reasonable to assume that impurities "similar" to the substance concerned will have a comparatively large influence on the rate of crystal growth, as they will more easily be incorporated into the growing crystals and destroy the regularity required for their orderly development.

For the case of TNT, the operativeness of structural factors has been proved in the experiments reported here. Pure TNT, or a mixture with the substance to be tested, was melted and cooled to 50°C in a capillary tube of 3.5 mm internal diameter. The capillary was held in a thermostat and the rate of linear crystal growth determined over a length of 20 cm. Table I gives the results; the "impurities" were added in approximately equimolecular quantities (0.02 mol. per mol. of TNT).

It is interesting to note that substances immiscible with molten TNT (β -phenylethyl acetate, ethyl palmitate) had no influence on the rate of crystallisation.

Clearly, substances which contain the 2, 4, 6-trinitrophenyl radical are the most active crystallisation depressants; the benzene nucleus or a 4-nitrophenyl radical have much less effect (No. 2, 3). Free hydroxyl compounds, even if they contain the 2, 4, 6-trinitrophenyl residue (4, 7), are less active than their esters, and among the latter there is a definite preference for aromatic as compared with aliphatic groups used to lock the free hydroxyl; saturated and unsaturated aliphatic radicals show the same effectiveness.

TABLE I.

| No. | "Impurity" | Weight (% of TNT) | Linear crystal growth (mm/min.) |
|-----|---|----------------------|--|
| 1. | None | — | 150 |
| 2. | Phenyl benzoate | 1.7 | 108 |
| 3. | 4-Nitrobenzyl acetate | 2.0 | 109 |
| 4. | 4-Nitrobenzyl benzoate | 2.1 | 102 |
| 5. | 2, 4, 6-Trinitrophenol (picric acid) | 1.9 | 99 |
| 6. | 2, 4, 6-Trinitrophenyl acetate | 2.25 | 80 |
| 7. | 2, 4, 6-Trinitrophenyl benzoate | 2.75 | 30 |
| 8. | 2, 4, 6-Trinitrophenyl- ethyl alcohol | 2.2 | 89 |
| 9. | 2, 4, 6-Trinitrophenyl- ethyl acetate | 2.5 | 28 |
| 10. | 2, 4, 6-Trinitrophenyl- ethyl butyrate | 2.75 | 26 |
| 11. | 2, 4, 6-Trinitrophenyl- ethyl palmitate | 4.2 | 24 |
| 12. | 2, 4, 6-Trinitrophenyl- ethyl methacrylate | 2.75 | 24 |
| 13. | 2, 4, 6-Trinitrophenyl- ethyl benzoate | 3.0 | 15 |
| 14. | Butyl 2, 4, 6-trinitro- benzoate | 2.6 | 43 |

It remains to be seen whether the combination of 2, 4, 6-trinitrophenyl radical and ester grouping ($-\text{COO}-$) is the only one active or whether other systems have a similar effect. The experiments are being continued in this direction; an attempt will also be made to differentiate between the effects of the "impurities" on the rate of formation of crystallisation nuclei and on the linear velocity of crystal growth, respectively.

This investigation has been carried out under the auspices of the Scientific Department, Ministry of Defence, and is published with its permission.

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POLAR CARBON-CARBON DOUBLE BONDS

It has been shown in recent years that lithium aluminium hydride (LiAlH_4) reduces only double bonds of the $\text{C}=\text{O}$ or $\text{C}=\text{N}$ type¹, but not $\text{C}=\text{C}$ double bonds in hydrocarbons², even in conjugated systems³. This may be ascribed to the fact that the active agent in these reactions is the negative hydrogen ion H^- in the form of a complex, which attacks the more positive end of the polar bond⁴.

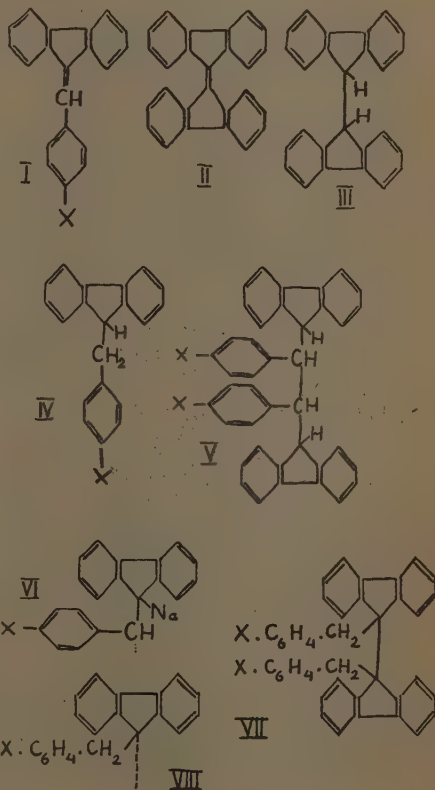
Pullman et al.⁵ and Coulson, Craig and Maccoll⁶ have shown that fulvenes, e.g. benzylidene-fluorene (I, $\text{X}=\text{H}$) and dibiphenylene-ethylene (II), have a central double bond of unusual character: it is polar, and the moment of (I) points towards the five-membered ring. This theory has been experimentally verified by Wheland and Mann⁷, Lumbroso, Pacault and Pullman⁸, and Bergmann and Fischer⁹.

It seemed, therefore, possible that such fulvene hydrocarbons would respond to the reducing action of lithium aluminium hydride. This is, indeed, the case. (II) was smoothly reduced in boiling ether to dibiphenylene-ethane (III), m.p. 247°C ¹⁰. p-Bromobenzylidene-fluorene (I, $\text{X}=\text{Br}$)¹¹ gave 9-(p-bromobenzyl)-fluorene (IV, $\text{X}=\text{Br}$), m.p. 151°C , which was synthesised for comparison by reaction of fluorenyl-lithium with p-bromobenzyl chloride¹².

A more intimate insight into the structure of the fulvenes was obtained in the case of the unsubstituted benzylidene-fluorene (I, $\text{X}=\text{H}$). In addition to 9-benzylfluorene (IV, $\text{X}=\text{H}$)¹³ of m.p. $130\text{--}131^\circ\text{C}$, a "dimeric" product was obtained ($2\text{I} + 2\text{H}$), which crystallized from acetic acid in needles of m.p. $204\text{--}205^\circ\text{C}$.

Two such dimeric products are theoretically possible, viz. (V) and (VII). (V) has been prepared by Schlenk and Bergmann¹⁴ by addition of sodium metal to (I, $\text{X}=\text{H}$) and subsequent hydrolysis of the addition product: the negative end of the polar $\text{C}=\text{C}$ bond responds preferentially to

the attack of the metal (Na), and the radical (VI) formed dimerises. (V) melts at 321°C and is, therefore, not identical with the "dimeric" product obtained in the reaction with lithium aluminium hydride. The "dimeric" product must, therefore, have formula (VII); indeed, it is identical with the 9,9'-dibenzyl-dibiphenylene ethane obtained by Fuson and Porter¹⁵ in an unambiguous synthesis. It is obvious that (VII) owes its formation to the following mechanism: the negative hydrogen ion attacks the positive end of the dipole and forms a radical (VIII), which dimerises. Thus, these experiments not only present evidence for the polar character of the central double bonds in (I) and (II), but



also for the direction of the moment predicted by the theory.

(*p*-Methoxybenzylidene-fluorene)¹⁸ (I, X=OCH₃) also gives two reaction products with LiAlH₄, 9-(*p*-methoxybenzyl)-fluorene of m.p. 109-110°^{14a, 18} and "dimeric" product, m.p. 218°C, which is different from (V, X=OCH₃) (m.p. above 300°C)^{14b} and is, therefore, most probably (VII, X=OCH₃).

A detailed report on this investigation will be published elsewhere.

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TWO CASES OF CIS-TRANS ISOMERISM IN THE SERIES OF DIBENZOFULVENE

It has been a generally accepted view that fulvenes, e.g. of the type (I), and related substances do not show the phenomenon of *cis-trans* isomerism (I,I')^{1, 2, 3}. This has been ascribed to the particular character of the central (semicyclic) bond in the fulvene system, which has recently been defined in terms of modern theory by Pullman *et al*⁴ and by Coulson, Craig and Maccoll⁵; the energy barrier between the two isomerides has been assumed to be very low.

In the course of an extended investigation, we have observed two cases of *cis-trans* isomerism in the dibenzofulvene series:

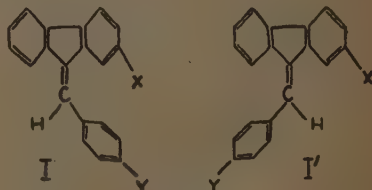


Fig. 1

A) X=NO₂, Y=NO₂

B) X=NO₂, Y=Br

(a) When 2-nitrofluorene was condensed with *p*-nitrobenzaldehyde in the usual manner (sodium ethoxide)⁶, a product

was obtained which could be separated into its constituents by means of boiling butyl acetate. The isomer *A* crystallised from this solvent in well-developed yellow needles, m.p. 251-252°C. (yellow melt). The isomer *B*, which was insoluble in butyl acetate, crystallised from chlorobenzene in orange-coloured flocculent fine needles, m.p. 287-288°C (red-brown melt).

Determination of the *dipole moments* of the two isomers (in benzene solution) has given the following figures :

$$(A) \mu = 7.85 \pm 0.1 \text{ Debye}$$

$$(B) \mu = 4.45 \pm 0.1 \text{ Debye}$$

The theoretical figures for the *cis* and *trans*-forms respectively, are 8 and 4 Debye, assuming a valency angle of 120° at the aldehyde carbon atom. (*A*) is the *cis*-, (*B*) the *trans*-isomer. The isomers are interconvertible with great ease. The *cis*-form, heated some degrees above the melting point, solidifies and melts again at 288-289°C (red-brown melt). Upon cooling, part crystallises at 260°C, the remainder at 230°C; if the solid mass is heated again, part melts at 235°C and the remainder at 288°C. The same phenomenon can be observed with a sample of the *trans*- compound, which has been heated for a few seconds above the melting point.

(b) The condensation product of 2-bromofluorene and *p*-nitrobenzaldehyde was fractionated by crystallisation from heptane into orange-coloured prisms of m.p. 154°C (*A*) and yellow needles of m.p. 168-169°C (*B*). Their dipole moments were (*A*) 5.30±0.05 Debye, (*B*) 3.80±0.05 Debye, while the theoretical figures are 3.5 and 5.5 Debye for the *trans* and *cis*-forms, respectively.

The *cis*-form (*A*), when heated above the melting point, rearranges itself quickly into the *trans*-form (*B*). As a matter of fact, the ensuing change in dipole moment can be used for a study of the kinetics of this isomerisation reaction.

The behaviour of the two pairs of substances shows the easy interconvertibility of the isomerides; the small difference in energy content is also expressed in the great similarity of the ultraviolet absorption spectra of each pair of compounds⁷. It appears that the nitro-group (*X*) in (*I*) raises the energy barrier between the isomerides somewhat, as in many other cases no indication of the existence of separate *cis-trans* isomers has been found.

However, it should be noted that Weisburger, Weisburger and Ray⁸ have recently described a pair of isomers in the case of 2,2'-di-(acetylamino)-dibiphenylene-ethylene (*II*), without proof, however, that the substances are geometrical isomers. It may also be recalled that Lugones⁹ observed that the condensation product (*III*) of *p*-dimethylaminobenzaldehyde with 2-nitrofluorene exists in three different easily interconvertible forms of m.p. 187°, 195° and 204°, respectively, which he considers crystallographically different, but not isomers. It is well known that the differentiation between these two concepts is sometimes difficult.

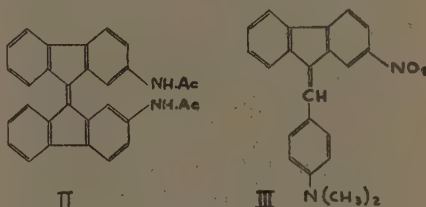


Fig. 2

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THE KINETICS OF POLYMERISATION OF METHACRYLIC ACID IN AQUEOUS SOLUTION.

As the products of a polymerization reaction are fixed in the molecular chain and are amenable to analysis, polymerisation reactions may be regarded as chemical "multipliers" for the investigation of free radical reactions. The behaviour of free radicals in aqueous solution is of interest both for inorganic and biological systems in an aqueous medium.

The polymerisation investigated was that of methacrylic acid in aqueous solution at 55-75°C, using hydrogen peroxide as catalyst. This polymerisation, in a sense, reflects the course of the decomposition of hydrogen peroxide into free hydroxyl radicals.

It was found that the overall reaction velocity was of the first order with respect to monomer concentration and was pro-

portional to the square root of the hydrogen peroxide concentration. A very remarkable dependence of the reaction velocity on the pH of the medium was observed. Fig. 1 gives the plots of $\log_{10} M_0/M$ against time for methacrylic acid polymerisations at different pH values (M_0 =initial conc. of monomer, M =monomer conc. at time t).

In Fig. 2 the kinetic constants k are plotted against pH:

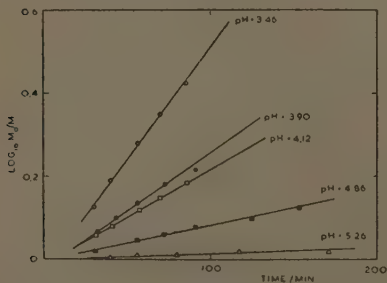


Fig. 1

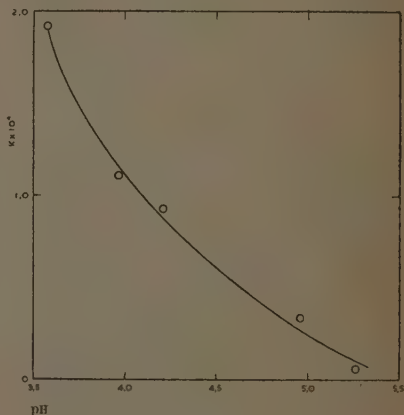


Fig. 2

This curve closely resembles the dissociation curves of monobasic acids and appears to demand the assumption that only the undissociated monomer is capable of interacting with free radicals, thus initiating and propagating the polymer chain. Indeed, at pH 5.5, where all of the monomer is ionized, the polymerisation velocity is practically zero under the operating conditions employed. If thus the k 's are only apparent constants, then the same — true constant k_0 of the polymerisation velocity should result from the ratios (k : corresponding fraction of undissociated monomers) at different pH. The data in Table I (α = degree of dissociation) show this assumption to be valid with reasonable approximation.

TABLE I.

| pH | $k \times 10^3 \text{sec}^{-1}$ | $1-\alpha$ | $K_0 = [k/(1-\alpha)] \times 10^3 \text{sec}^{-1}$ |
|------|---------------------------------|------------|--|
| 3.58 | 19.2 | 0.780 | 24.6 |
| 3.97 | 11.1 | 0.592 | 18.7 |
| 4.21 | 9.31 | 0.455 | 20.5 |
| 4.96 | 3.36 | 0.114 | 29.5 |
| 5.26 | 0.58 | 0.057 | 10 |

$$k_0 = 2.33 \pm 0.42 \times 10^{-4} \text{sec}^{-1}$$

From the degree of polymerisation and its velocity, one determines the order of magnitude of the constant k_i of the initiation velocity, *i.e.* the velocity of decomposition of hydrogen peroxide into free radicals.

A detailed account of this investigation will be published elsewhere.

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*An abstract of a thesis submitted by G. Blauer to the Hebrew University, in partial fulfilment of the requirements for the degree of Doctor of Philosophy.

THE CAUSE OF THE THERMOCHROMY OF BIANTHRONE

Bianthrone (I), upon heating, turns reversibly dark-green¹. This effect has been related by E. Bergmann² to the inability of (I) to arrange itself in one plane. The four *ortho*-hydrogen atoms interfere with each other, so that the two halves of the molecule must be inclined against each other. In this state, obviously, the central linkage cannot be a real double bond (the four substituents of which must be in one plane). It can then be assumed that at higher temperatures this central bond stretches sufficiently to decrease the mutual interference of the *ortho*-hydrogen atoms. This explanation is, at least partly,

supported by the experiments of Grubb and Kistiakowsky³ on the temperature dependence of the absorption spectrum of (I).

This theory leads to the following experimentally verifiable conclusion: if one introduces in *ortho*-position to the central double bond substituents larger than hydrogen, the stretching of the central bond would not suffice to permit the substituents to arrange themselves in one plane, even at higher temperature. It should thus be possible to destroy the thermochromy. *This effect has now been observed.* 4,4'-Dimethyl-, 4,4'-dibromo- and 4,4'-dimethoxy-bianthrone (II) are no longer thermochromic. Their spectra at ordinary temperature do not substantially differ from those of 4-methyl-, 4-bromo- and 4-methoxy-anthrone (III), respectively (Fig. 1, 2).

The anthrones (III) were obtained by cyclisation of the corresponding substituted 2- or 2'-benzyl-benzoic acids (IV) and the latter, by suitable reduction of the keto-acids (V). These were prepared by the reaction of 2-methyl- and 2-methoxy-phenylmagnesium bromide with phthalic anhydride⁴, and — for the bromo-derivative — by Friedel-Crafts reaction of 3-bromophthalic anhydride and benzene⁵. The anthrones (III) were oxidised by ferric chloride in acetic acid to the bianthronyls (VI) and these successively enolized with alkali in anhydrous alcohol (to VII) and dehydrogenated with quinone⁷ in acetone solution to (II).

The characteristics of the substances concerned are summarized in the following Table:

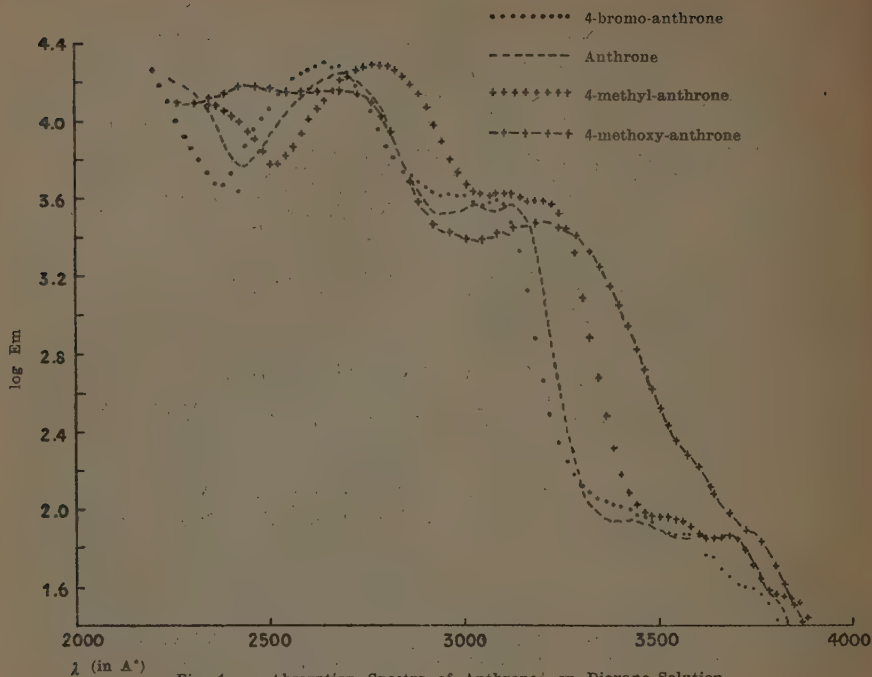


Fig. 1. — Absorption Spectra of Anthrones on Dioxane Solution.

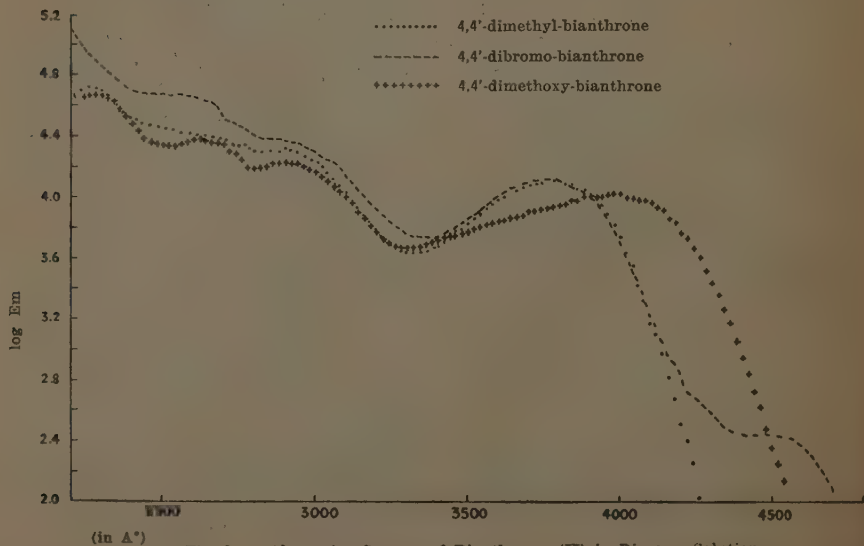
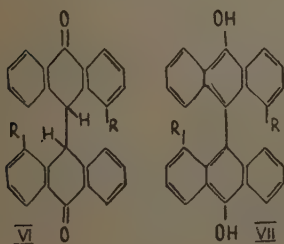
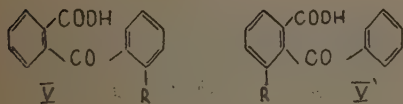
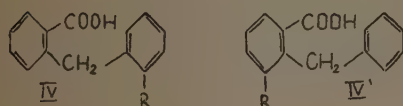
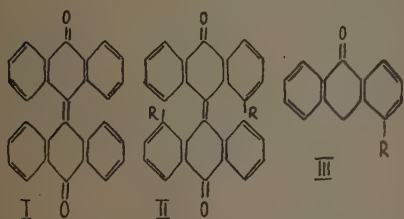


Fig. 2. — Absorption Spectra of Bianthrone (II) in Dioxane Solution.

TABLE

| | |
|---|--|
| 2-(2'-Methyl-benzoyl)-benzoic acid ⁷ | m.p. 128-130°; 90% yield |
| 3-Bromo-2-benzoyl-benzoic acid | m.p. 225-226°; 96% yield |
| 2-(2'-Methoxy-benzoyl)-benzoic acid | m.p. 141-144°; 50% yield |
| 2-(2'-Methyl-benzyl)-benzoic acid | m.p. 128-128.5; 95% yield |
| 3-Bromo-2-benzyl-benzoic acid | m.p. 150.5-151.5° (from benzene-heptane); 22% yield (in two stages) |
| 2-(2'-Methoxy-benzyl)-benzoic acid | m.p. 112-113°; 95% yield |
| 4-Methyl-anthrone ⁸ | m.p. 126°; 80-90% yield |
| 4-Bromo-anthrone | m.p. 133.5° (from heptane or alcohol); 80-90% yield |
| 4-Methoxy-anthrone ⁹ | m.p. 130-131°; 30-40% yield |
| 4,4'-Dimethyl-bianthranyl | m.p. 233° (from ethyl-cellosolve); 40% yield |
| 4,4'-Dimethyl-bianthrone | m.p. 330-335° (from xylene); 75% yield |
| 4,4'-Dibromo-bianthranyl | m.p. 220-221°; quantitative yield |
| 4,4'-Dibromo-bianthrone | m.p. 340° (from xylene); 50% yield |
| 4,4'-Dimethoxy-bianthranyl | (not isolated in pure form); 75% yield |
| 4,4'-Dimethoxy-bianthrone | m.p. 343-345° (from xylene); 70% yield |



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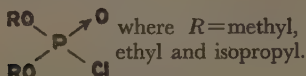
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THE NUCLEOPHILIC SUBSTITUTION OF HALOGEN IN ALKYL HALOPHOSPHONATES

As a part of our study of the properties of phosphorus-halogen bonds, we have investigated the mechanisms of the nucleophilic halogen substitution in three alkyl chlorophosphonates:



The reactions studied were (a) the solvolysis in various ethanolic media, (b) the reaction with ethoxide ions in absolute ethanol, (c) the reaction with fluoride ions

in ethanolic solvents, (d) the reaction with di-n-butyl amine in absolute ethanol.

The reactions with ethoxide and fluoride ions and with di-n-butyl amine were found to proceed by second order kinetics and are therefore bimolecular.

The relatively small solvent and salt effects on the rates of the solvolytic reactions, together with the absence of any considerable effect of silver ions (see Table III) led us to suppose that in the solvolytic reactions too, the reaction is bimolecular with ethanol and water molecules acting as nucleophilic reagents. This fact permitted us to introduce (Table II) a direct comparison of the reaction with ethanol and the reactions with other nucleophilic reagents.

TABLE I

*The solvolysis of alkyl chlorophosphonates in ethanolic media.
First order rate constants $\times 10^4 \text{ sec.}^{-1}$.*

| chlorophosphonate | Temp. °C | "100%" ethanol | "90%" ethanol | "80%" ethanol | "60%"* ethanol |
|-------------------|----------|----------------|---------------|---------------|----------------|
| Diisopropyl | 0 | 0.097 | — | — | 0.63 |
| | 10.08 | 0.187 | 0.64 | — | — |
| | 25.15 | 0.61 | 2.03 | 3.12 | 5.72 |
| Diethyl | 0 | 0.23 | — | 1.82 | 2.81 |
| | 10.08 | 0.54 | — | 3.77 | — |
| | 25.15 | 1.68 | — | 12.8 | — |
| Dimethyl | 10.08 | 1.6 | — | 9.9 | — |

* "X%" ethanol is the solvent obtained by mixing X volumes of specially dried ethanol with 100-X volumes of distilled water.

TABLE II

Rates of substitution reaction of di-isopropyl and diethyl chlorophosphonate at 0°C with various reagents in absolute ethanolic solution.

| Second order rate constant | | litre mol. sec. |
|----------------------------|-----------------------|-----------------------|
| Reagent | Di-isopropyl | Diethyl |
| O Et ⁻ | 0.011 | 0.26 |
| F ⁻ | 0.031 | 0.28 |
| di-n-butyl amine | 0.0015 | — |
| EtOH | 5.58×10^{-7} | 1.31×10^{-6} |

TABLE III

Effect of added reagents on the solvolysis of di-isopropyl chlorophosphonate at 25°C in "60%" ethanol.

| Reagent added | First order rate constants in sec. ⁻¹ |
|--------------------------|--|
| none | 5.72×10^{-4} |
| 0.1 N KNO ₃ | 6.2×10^{-4} |
| 0.01 N AgNO ₃ | 5.68×10^{-4} |

We also found that the solvolysis of di-isopropyl chlorophosphonate, unlike that of the corresponding fluoride, is not subject to acid catalysis.

The great difference between the rate of reaction with nucleophilic reagents and of the solvolytic reaction enabled us to ignore, in studying the former, any contributions from the simultaneous slow solvolysis.

It is interesting to note that since the reaction with F^- proceeds so much faster than the solvolysis, even in aqueous media, the product of reaction is essentially pure dialkyl fluorophosphate.

The products of the reactions have been isolated and found to confirm the bimolecular mechanism. A number of these products are new substances; these include di-isopropyl ethyl phosphate and di-isopropyl di-n-butyl phosphamide.

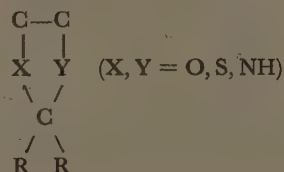
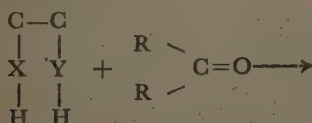
The Arrhenius parameters of the reactions studied have been determined and the dependence of activation energy and entropy terms upon the nature of the alkyl group R established. Pending a fuller description of our results elsewhere, we may state that both activation energy E and the term PZ of the Arrhenius equation are low, E ranging between 11-14 Kcal/mole and PZ between 10^5 - 10^7 lit/mol. sec.

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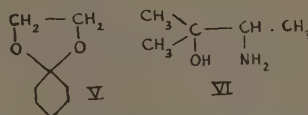
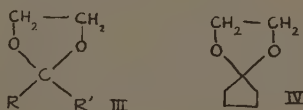
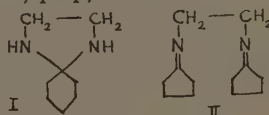
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ON SOME UNEXPECTED REACTIONS OF CYCLOPENTANONE AND METHYLISOBUTYLKETONE

In the course of an investigation of the synthesis of heterocyclic substances according to the general formula



and of their properties, it was found that cyclopentanone and methylisobutylketone frequently behave abnormally. This becomes particularly clear if one compares them, for example, with cyclohexanone and methylpropylketone.



1) With ethylenediamine, cyclohexanone gives an imidazolidine (I), cyclopentanone (and aliphatic ketones), the double Schiff's base (II), even if the reagents are used in molar ratio or if an excess of the diamine is employed.

2) In the infra-red spectrum of a 2,2-disubstituted 1,3-dioxolane (III), there appear four characteristic bands in the 1030-1200 cm^{-1} region, one of them, which usually has the highest extinction coefficient, at 1063-1098 cm^{-1} . For reasons which there is not space to discuss here, it probably belongs to the following vibration:



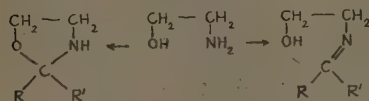
This band is shifted in the methylisobutyl-

ketone derivative (III, $R=CH_3$, $R'=iso-C_4H_9$), and in (IV), as the following figures show :

| | |
|-----------------------|-------------------------------------|
| 2, 2-diethyl | 1077 ($\epsilon=2.5 \times 10^4$) |
| 2-methyl-2-isobutyl | 1101 ($\epsilon=3 \times 10^4$) |
| 2-tetramethyleno (IV) | 1107 ($\epsilon=2 \times 10^4$) |

In the dipole-moments², the peculiar position of the derivatives of methylisobutylketone and cyclopentanone is also expressed. A regular dioxolane ring should have the moment 1.1 and both 2, 2-diethyl-1, 3-dioxolane and the cyclohexanone derivative (V) have the expected moment. (IV) has a moment of 1.24 ± 0.02 D, and the 2-methyl-2-isobutyl compound 1.17 ± 0.02 D. This can be interpreted as due to a deformation of the heterocyclic system.

3) The most unexpected results were obtained in the condensation with amino-alcohols. It can be stated that saturated aliphatic ketones give, with ethanolamine, oxazolidines, aromatic aldehydes and ketones and α, β -unsaturated carbonyl compounds, preferentially Schiff's bases, according to the following scheme :



Substitution at the carbon atoms of the ethanolamine favours formation of the ring compound, even for aromatic aldehydes and ketones. Methylisobutylketone as well as diisobutylketone give, with ethanolamine, mixtures which according to the molecular refraction contain 60% and 75%, respectively, of the open form. This is shown in the Table.

The Table also shows that cyclopentanone and ethanolamine give largely a Schiff's base. In all cases, this abnormal tendency is suppressed in the reaction with the amino-alcohol (VI).

The same result was obtained by an analysis of the infra-red spectrum of the "oxazolidines", taking into account that

the $\begin{array}{c} O \quad N \\ \backslash \quad / \\ C \end{array}$ system has a characteristic

three-band absorption in the 1100-1200 cm^{-1} region, while the $C=N$ double bond absorbs in the 1620-1670 cm^{-1} region.

No explanation for these anomalies is offered. It is unlikely that the effect is of steric nature; in such case, there should be no marked difference in the behaviour of cyclopentanone or cyclohexanone — nor should pinacolone behave normally; there should be much more interference.

The following facts may be recalled to show that there are as yet unknown factors influencing the behaviour of methylisobutylketone. Both with glycol³ and with (VI), the reaction velocity of this ketone is extremely low; of all aliphatic ketones, methylisobutylketone is the least reactive towards chloroform⁴, and its condensation with formaldehyde is a slow and unsatisfactory reaction.

There are also a number of known chemical differences between the cyclopentane and cyclohexane systems; they have been discussed by F. Bergmann and A. Weizmann⁵. Cyclopentanone has a normal molecular refraction ($n_D^{30}=1.4366$; $d_4^{30}=0.9480$; MR [found] 23.17; MR [calc.], 23.11), but in its infra-red spectrum, the carbonyl band lies at an abnormal wave-length (1740 instead of 1720 cm^{-1})⁶. The difference in molecular structure between cyclopentene and cyclohexene⁷ is also significant in this respect.

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TABLE

| Amino-alcohol | Condensation product of Ketone | Mol. Refr. (obs.) | Mol. Refr. (calcd.) | |
|-------------------------------------|-----------------------------------|-------------------|---------------------|-------------|
| | | | Schiff's Base | Oxazolidine |
| ethanolamine | methylisobutylketone | 43.07 | 43.70 | 42.20 |
| ethanolamine | diisobutylketone | 57.14 | 57.56 | 56.06 |
| ethanolamine | cyclohexanone | 39.55 | 41.50 | 40.00 |
| ethanolamine | cyclopentanone | 36.13 | 36.76 | 35.26 |
| ethanolamine | pinacolone | 42.23 | 43.70 | 42.20 |
| 2-amino-3-methyl- 3-butanol (VI) | dipropylketone | 60.07 | 62.16 | 60.66 |
| " | methylisobutylketone | 55.74 | 57.54 | 56.04 |
| " | diisobutylketone | 69.51 | 71.39 | 69.89 |
| " | cyclohexanone | 52.62 | 55.36 | 53.86 |
| " | cyclopentanone | 48.96 | 50.72 | 49.22 |
| " | pinacolone | 55.34 | 57.54 | 56.04 |

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2-cyclohexenone (I) which has recently been made available¹, had to be substituted, at the marked* position by a radical, capable of forming the bridge in the desired phenanthrene system, and still retaining reactive groups on which further transformations could be based.

A satisfactory solution of the problem lies in the observation that (I) smoothly undergoes Michael condensation with compounds containing active methylene groups, giving adducts of type (II) in high yield, e.g. (IIa) and (IIb) with diethyl malonate and ethyl cyanoacetate, respectively. 2-Phenyl-2-cyclohexenone was used as a model substance in the study of this condensation.

Some of the data pertinent to the adducts obtained are given in Table I.

As an example of the conversion of such adducts to an octahydrophenanthrene derivative, the following scheme is characteristic:

THE MICHAEL CONDENSATION OF 2-ARYL-2-CYCLOHEXENONES

In a study aimed at the synthesis of pharmacologically active compounds related to morphine, the 2-(2', 3'-dimethoxyphenyl)-

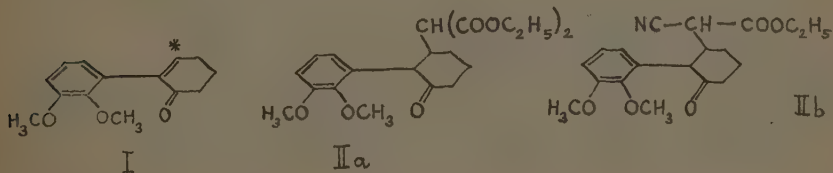
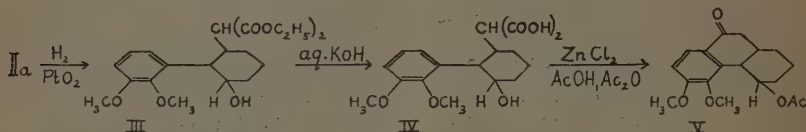


TABLE I.

| Aryl of 2-aryl-2- cyclohexenone | Adduct with | Yield (%) | M.p. (°C) | Recrystal- lised from | M.p., DNP* (°C) | Recrystal- lised from |
|---------------------------------------|---------------------|--------------|--------------|--------------------------|-----------------------|------------------------------|
| phenyl | diethyl malonate | 96 | oil | — | 119-120 | alcohol |
| phenyl | nitromethane | 80 | 126.5-127.5 | dioxan- alcohol | 155-157 | alcohol- chloroform |
| 2, 3-dime- thoxyphenyl | dimethyl malonate | 97 | oil | — | 162-163 | alcohol- ethyl acetate |
| " | diethyl malonate | 94 | oil | — | 117-119 | alcohol |
| " | methyl cyanoacetate | 95 | 114 | butanol | 167 | alcohol- chloroform |
| " | ethyl cyanoacetate | 90 | 95 | butanol | 176-178 | alcohol- chloroform |

(*DNP=2,4-Dinitrophenylhydrazine)



(IIa) is hydrogenated in alcohol using platinum oxide as catalyst. The diethyl [3-hydroxy-2-(2', 3'-dimethoxyphenyl)-cyclohexyl]-malonate (III) is an oil, yielding a crystalline acetate, m.p. 83-84°C (from methylcyclohexane).

(*Anal. Calc.* for $C_{21}H_{30}O_7$: C, 63.8; H, 7.8. Found: C, 63.6; H, 7.9).

Its alkaline hydrolysis gave the corresponding dicarboxylic acid (IV), m.p. 185°C (dec.), (from dioxan-methylcyclohexane). (*Anal. Calc.* for $C_{17}H_{22}O_7$: C, 60.3; H, 6.5. Found: C, 59.7; H, 6.5). Cyclisation of the acid by means of zinc chloride-acetic acid-acetic anhydride, was accompanied by decarboxylation, and 3, 4-dimethoxy-5-acetoxy-10-keto-5, 6, 7, 8, 9, 10, 13, 14-octahydrophenanthrene (V) was formed. It was recrystallised from ethanol and melted at 129°C. (*Anal. Calc.* for $C_{18}H_{22}O_5$: C, 67.9; H, 6.9. Found: C, 67.7; H, 6.9).

Its saponification yielded the corresponding 5-hydroxy-compound, m.p. 97.5-

98°C (from methylcyclohexane-benzene). (*Anal. Calc.* for $C_{16}H_{20}O_4$: C, 69.6; H, 7.3. Found: C, 69.5; H, 7.3).

Details of this investigation will be published elsewhere.

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THE EFFECT OF AMMONIA-NITROGEN AND HYDROGEN SULPHIDE ON THE BIOCHEMICAL OXYGEN DEMAND OF SEWAGE

In a study of the efficacy of a newly-built Imhoff tank in the Haifa Bay area, the 5-day biochemical oxygen demand (B.O.D.) at 20°C was found to be the same in the effluent as in the influent. Compared

to the influent, the effluent showed an unsatisfactory decrease in suspended solids (33%), an increase in ammonia-nitrogen, and, judging by its odour and dark colour, an increase in sulphides.

Obviously, the decrease in suspended solids should have brought about a corresponding decrease in B.O.D. However, in the light of the literature¹⁻¹⁰ and the afore-mentioned observations and analyses, it was assumed that the anticipated decrease was compensated for by a B.O.D. enhancing effect due to ammonia-nitrogen and hydrogen sulphide. The assumption was tested in the experiments to be described.

These consisted chiefly in taking raw domestic sewage, varying its ammonia-nitrogen and hydrogen sulphide content, and determining the resultant B.O.D. Three samples of sewage were taken, each of which formed the basis of one series of experiments. The source of ammonia-nitrogen employed was ammonium chloride.

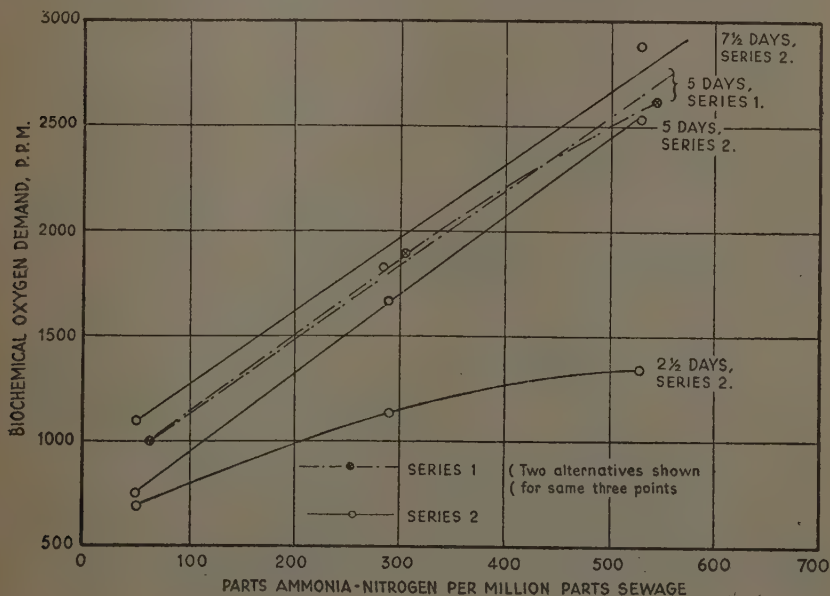
In figure 1 the B.O.D. of sewage is shown to be directly proportional to the

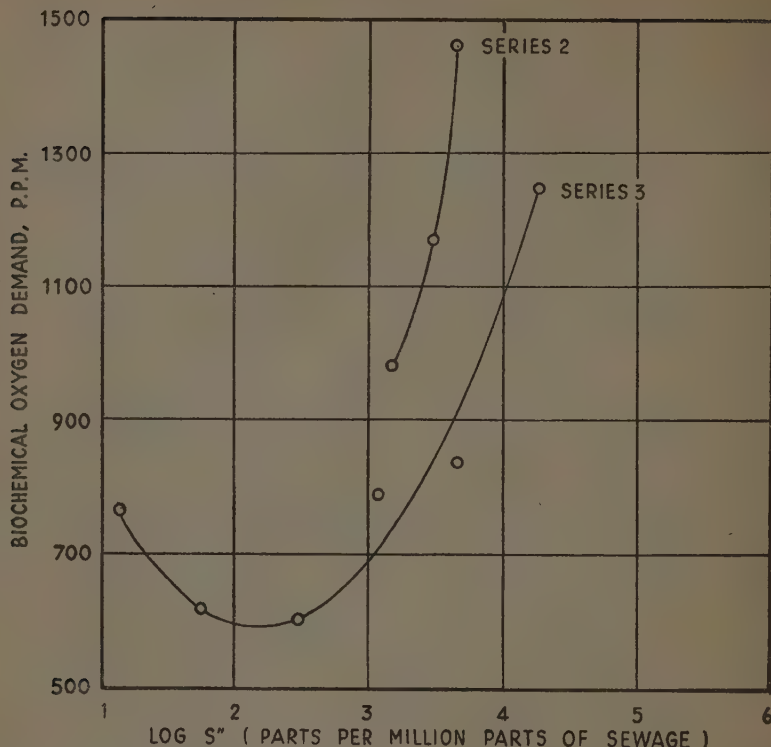
concentration of ammonia-nitrogen. Except for the 2½-day period, the slopes for different series of different time-periods are almost identical, and the relationship is practically linear.

The effect of hydrogen sulphide is shown in figure 2, where the 5-day B.O.D. is plotted against log S". As the concentration of S" increases, the B.O.D. first decreases, then passes through a minimum, and subsequently rises until it exceeds the original value of the sewage.

When both ammonia-nitrogen and hydrogen sulphide were added, the latter at a concentration which would leave the B.O.D. unaffected, the B.O.D. of the sewage nevertheless decreased.

It appears that ammonia-nitrogen produces an increased growth and multiplication of the sewage bacteria, thus increasing the B.O.D. As to the effect of hydrogen sulphide, the repressive effect on the B.O.D. is probably due to a repression of bacterial activity. It remains to be seen whether the enhancing effect of higher concentrations is due to inorganic ox-





dation, biochemical oxidation, or both.

Further details will be supplied at a later date.

This note is based on a research project sponsored by the Research Council of Israel and conducted under the guidance of Prof. J. Breuer.

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PRELIMINARY STUDIES ON FLAX RETTING IN ISRAEL

The retting of flax and other vegetable fibres is of great economic importance for this country, particularly in view of the urgent need to develop local industries.

High-grade fibres depend on: good varieties of fibre plants, suitable micro-organisms and good retting methods.

Fibre plants are now being grown in various parts of the country including the Negev area, and varieties suitable for different soils are being investigated. Experimental tank retting of flax under natural conditions i.e. with their natural microflora, is proceeding satisfactorily, giving gratifying results. The quality of the retted fibre obtained is sufficiently good for larger scale retting operations.

The natural retting flora on the straws and in the retting tanks is being investigated in order to obtain, if possible, the best retting organisms which yield a superior grade quality fibre. A preliminary examination of the flora from retting liquors from four different varieties of flax grown in the Kubeiba district revealed the presence of a number of bacteria common to all four samples. Besides cocci, short rods and small sporing forms, a striking feature was the predominance of clostridial forms (see Plate 1). At least three different types of clostridia were readily distinguished, and subsequently isolated in pure culture: a plectridial anaerobe exhibiting long slender tennis-racket-like sporangia, an orange-pigmented anaerobe, and a short, stout facultative anaerobe. An orange-pigmented, aerobic, non-sporing, short rod, producing a pleasant fruity odour was also one of the major organisms found in each retting liquor sample.

The finding of plectridial and orange pigmented clostridia in Israel retting liquors is of special interest because of their reputed role as the causal agents of the retting of industrial vegetable fibres. Friebes and Winogradsky (1895) were the

first to describe the remarkable flax retting activity of a plectridial anaerobe isolated by Friebes. A similar if not identical plectridial form was isolated by Störmer (1903) in Germany, by Beijerinck and van Delden (1904) in Holland and later by Ruschmann and Bavendamm (1925) in Germany. All of them strongly supported Friebes and Winogradsky, who maintained that the plectridial anaerobe named *Cl. pectinovorum* (Synonym *Plectridium pectinovorum* Störmer) was the true causal agent of retting. Carbone and Tombolato (1917) were the first to isolate a yellow-orange clostridium, their *Clostridium felsineum*, from Italian hemp retting pits and proved its superior abilities in the industrial retting of various fibre plants. In more recent years, the high grade fibre produced in Belgium is attributed to the presence of large numbers of the orange *Cl. felsineum*. Lanigan (1950) believed that in the early development of the retting industry in Australia, the low quality fibre obtained under natural retting conditions was due to the lack of this yellow-orange clostridium, and succeeded in improving their fibres by introducing heavy inocula of this organism in their retting tanks. As far as we know there is no mention in the literature of *Cl. pectinovorum* or closely related plectridia, nor pigmented clostridia occurring in the microbial flora in the retting industry in Great Britain (Allen-1946 a, b).

A preliminary attempt was made to investigate the retting abilities of some of the organisms isolated from the first samples of retting liquors. This experiment was merely intended as a feeler for a more serious study of the micro-biology of retting, and was limited in its scope. The results however are of sufficient interest to warrant their publication. The experiment was conducted as follows:

Method. 100 gr. dried flax plants were placed in each of nine long glass tubes, 120 cm in length and 5 cm in diameter, and plugged with cotton wool. Eight of these tubes were steam sterilised for $\frac{1}{2}$ hr.

on each of three successive days. The ninth tube was left unsterilised. 120 ml of young active cultures of a single or combined isolate (60 ml culture on 1% tryptone plus 0.2% glucose plus 0.025% sodium thioglycollate, and 60 ml culture on 10% flax extract plus 0.2% glucose) were poured into seven of the tubes. 120 ml of the same but uninoculated medium were poured into the remaining two tubes; 120 ml of a natural retting liquor were added as retting culture to the tube containing the sterilised flax, whereas the

ninth tube with the natural bacterial flora on the non-sterilised flax served as a control. Sterile water up to 1,200 ml total liquid volume was carefully added to each tube, and the tubes were incubated at 32°. Some of our observations on the retting liquors and the retted straws after four days, are given in the table below. The straws were dried at 45° without previous washing. The grading of the retted fibres evaluated by several persons with expert knowledge of flax fibres, is included in the table.

Experimental data on flax retted for four days at 32° with bacteria isolated from local retting liquors

| Tube No. | Culture | Retting liquor | | | Wt. of dry retted straws* (gr) | Fibre yield | | Grading** |
|------------------------------------|-------------------------------|----------------|--------|---|----------------------------------|-------------------------|--|--------------------|
| | | Final pH | Colour | Odour | | 60 gr dry retted straws | 100 gr non-retted straws | |
| 1 | Hf 1 orange clostridium | 4.45 | YBR | acidic | 85.5 | 11.2 | 16.0 | Premier-textile |
| 2 | Hf 2 plectridium | 4.57 | YB | slightly aromatic | 95.5 | 8.52 | 13.6 | Premier-twine |
| 3 | Hf 3 orange bacterium | 4.6 | YB | aromatic | 79 | 8.52 | 11.2 | do. |
| 4 | Hf 1+2 | 4.52 | BR | slightly aromatic | 81.5 | 12.12 | 16.5 | do. |
| 5 | Hf 1+3 | 4.49 | BR | very slightly aromatic | 76.5 | 8.54 | 10.9 | Second twine |
| 6 | Hf 2+3 | 4.57 | | slightly aromatic | 83 | 7.92 | 11.0 | do. |
| 7 | Hf 1+2+3 | 4.46 | BR | aromatic | 84 | 7.56 | 10.6 | Below second twine |
| 8 | Retting liquor | 4.53 | YB | slightly aromatic, slightly putrid | 83 | 10.09 | 13.9 | do. |
| 9 | Natural flora | 4.9 | YB | acidic, aromatic | 89.5 | 12.44 | 18.7 | Premier-textile |
| Y — Yellow B — Brown R — Red | | | | | *100 gr non-retted straws taken. | | **Opinion of several local flax experts, based on colour, handle, strength and fibre dimensions. | |

PLATE I & II

Microphotographs of smears of retting liquors from 4 varieties of flax grown in the Kubeiba district, showing large numbers of various types of costridial organisms and free spores (x1550).

A



B



PLATE I

C



D



PLATE II

The results demonstrate that the natural flora, consisting mainly of pigmented and non-pigmented anaerobic clostridia and a pigmented aerobic bacterium, produces a good quality fibre within four days. The same result was obtained with a pure culture of the orange coloured clostridium. We note that the pH value of all the retting liquors fell to 4.5–4.9, due to the production of organic acids.

This is of interest as a low pH value of the retting liquor is generally considered to have a somewhat harmful effect on the fibres.

In conclusion, the above is a preliminary note on an investigation which is being conducted on the local microbial flora and suitable systems of retting of vegetable fibres of economic importance.

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DIFFERENCES IN CHEMICAL ACTIVITY BETWEEN THE CLOSTRIDIAL AND ROD STAGES OF CLOSTRIDIUM BUTYLICUM

The so-called clostridial stage of the butyl alcohol and butyric acid bacteria has been regarded as a pre-spore form which accumulates reserve material, and is not active in the various steps of the fermentation process. In the case of *Cl. acetobutylicum* (Weizmann), it has been definitely stated that "the vegetative cells are the form which bring about the chemical changes in the medium"¹.

The peculiar properties of a strain of *Cl. butylicum*, isolated from Jerusalem soil, made it possible to investigate this problem. At a certain stage of the fermentation, this strain forms large masses of a polysaccharide (*see plate opp. page 113*) which can be easily separated from the fermenting fluid by decantation. By treatment in a Waring Blendor, this polysaccharidic material gives a cell suspension which consists almost entirely of clostridial forms.

Suspensions of vegetative cells, free from clostridia, were obtained from young cultures (not older than 10 hours) in the usual manner.

The biochemical activities of the two types of suspensions were compared, in particular their ability to reduce acetone to isopropyl alcohol. The experiments showed that the clostridial suspensions were much more active in this respect than the vegetative forms, and that they carry out the reduction process at 30°, but not at 37°C.

The clostridial form differs from the vegetative one also in its ability to form the above-mentioned extracellular polysaccharide. This is very conspicuous even in the case of suspensions of resting cells. The polysaccharide appears at first in the form of enormous capsules surrounding the individual clostridial cells; later on, these capsules coalesce to form the mass described above. A certain amount of po-

lysaccharide can even be formed without the presence of carbohydrate in the suspending fluid, as clostridia showed this phenomenon also in distilled water. In this case, iodine-staining material contained in the clostridial cells disappears gradually, so that it can be assumed to be the source of the extracellular polysaccharide.

As a result of these experiments it can be concluded that clostridial forms are biochemically active and can by no means be considered as "inert".

A detailed report on this investigation will be published elsewhere.

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THE ACTION OF SOME WATER-SOLUBLE POLY- α -AMINO ACIDS ON BLOOD CLOTTING

Certain high molecular weight materials found in nature, such as heparin and protamine, are known to decelerate blood clotting¹. The similarity between the natural protamines and the synthetic basic water-soluble poly- α -amino acids², encouraged us to examine the action of the latter on blood clotting.

In experiments carried out *in vitro* with human blood, it was found that the basic poly- α -amino acids, poly-L-lysine, poly-DL-lysine³ and poly-DL-ornithine⁴, cause a marked prolongation of clotting time at a concentration of 100 μ g per ml. of blood. On the other hand, the neutral poly-DL-alanine⁵ and the acidic poly-aspartic acid⁶ and poly-D-glutamic acid⁷ do not retard clotting of human blood at the same concentration.

For a detailed study of the mode of anticoagulant action of the basic poly-amino acids, poly-L-lysine was chosen. It has been shown that its clot-retarding effect is, in the concentrations used, not due to the inactivation of fibrinogen, nor to the augmentation of antithrombic activity of the blood. By making use of the two-stage method for the determination of prothrombin^{8,9}, it could be demonstrated that the clot retardation induced was associated with the presence of elevated prothrombin levels in the serum of the slowly clotting blood. This finding indicates that poly-L-lysine interferes with the formation of thrombin from prothrombin in the clotting blood.

Thromboplastin acts as a catalyst in prothrombin conversion; it could be assumed, therefore, that it is inactivated by the basic poly-amino acids. Experiments with thromboplastin from rabbit brain proved, indeed, that it is partially inactivated on incubation with poly-lysine. The basic poly-amino acids form precipitates with cephalin in aqueous solution; as thromboplastin contains a phospholipid component¹⁰, it seems likely that its inactivation by the basic poly-amino acids is caused by the reaction of the latter with the phospholipid component.

The basic poly-amino acids show strong antiheparinic activity, in spite of their decelerating effect on blood clotting. Poly-lysine and poly-ornithine, in suitable concentrations, may neutralize the clot retarding effect of heparin, and restore the clotting time of heparinized blood to normal. The neutralization of heparin by the basic poly-amino acids is probably due to the neutralization of the acidic sulfonic groups of heparin by the basic amino group of poly-lysine and poly-ornithine, and the formation of an inactive poly-salt.

In their ability to neutralize heparin and in their mode of blood anticoagulant action, the basic poly-amino acids closely resemble protamine. It is thus believed that the synthetic basic poly-amino acids

may find some clinical use as antiheparins *in vitro* and *in vivo*.

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THE ACTION OF SOME WATER-SOLUBLE POLY- α -AMINO ACIDS ON BACTERIA

Some of the antibiotics formed by fungi and bacteria have been found to consist of α -amino acids linked by peptide bonds¹. The recent work of Bloom, Watson, Cromartie and their associates² showed that even the cells of higher animals may, in certain cases, e.g. in the infection with anthrax, excrete antibacterial substances of peptide nature.

These facts encouraged us to study the biological properties of some *synthetic* water-soluble poly- α -amino acids, which

were synthesized in this laboratory³. The water-soluble poly- α -amino acids investigated may be classified, according to their electrochemical properties, into neutral, acidic and basic poly-amino acids. In the present study, poly-DL-alanine⁴ was chosen as representative of the neutral poly-amino acids (it is the only known water-soluble representative of this group); the acidic poly- α -amino acids are represented by synthetic poly-aspartic acid⁵, and the natural poly-D-glutamic acid⁶, and the basic ones by poly-L-lysine and poly-DL-lysine⁷.

The determination of the bacterial spectrum of the various polymers, studied by the usual technique, proved that the neutral and acidic poly-amino acids have no antibacterial activity; the basic poly-amino acids, on the other hand, show strong antibacterial activity against both Gram-positive and Gram-negative bacteria. Poly-L-lysine, for example, inhibits the growth of *Micrococcus pyogenes* var. *aureus*, *Escherichia coli* and *Bacillus anthracis*, in synthetic and semi-synthetic media at a concentration of 1-2 μ g, 5 μ g, and 10 μ g, per ml., respectively.

At concentrations leading to growth inhibition, the basic poly-amino acids agglutinate living bacteria as well as bacteria killed by heating. The basic polymers also agglutinate human red blood cells and those of rabbits and sheep. At a concentration range of 1-50 μ g per ml. no haemolysis of bacteria or red cells was observed.

The interaction of the basic poly-lysine with bacteria seems to be reversible; thus it is possible to restore the growth of *E. coli*, previously incubated with the above basic polymers, by the addition of equimolar concentrations of the acidic poly-amino acids, poly-aspartic acid and poly-glutamic acid. As it has been shown in other experiments that poly-lysine in aqueous solution forms insoluble poly-salts with poly-aspartic and poly-glutamic acids, it is reasonable to formulate the following scheme for the growth restora-

tion, by means of poly-aspartic acid, of bacteria incubated with poly-lysine:

Bacterium + poly-lysine = (Bacterium \longleftrightarrow poly-lysine).

(Bacterium \longleftrightarrow poly-lysine) + poly-aspartic acid = (bacterium + (poly-lysine \longleftrightarrow poly-aspartic acid)).

Most of our experiments were carried out at pH 7.4. At this pH, most of the bacteria studied are negatively charged, the basic poly-amino acids positively, and the acidic poly-amino acids also negatively. The conclusion offers itself that at least part of the attraction between the bacteria and the antibacterial polymers, as between the basic and acidic poly-amino acids, is caused by electrostatic forces.

In respiration experiments carried out with the Warburg manometric technique, it was found that the addition of poly-L-lysine to *E. coli* and *M. aureus*, using glucose as a substrate, caused reduction in respiration. The oxygen uptake decreased with increasing concentration of the polymer and with increasing time of incubation.

The basic poly-amino acids resemble; therefore, in their antibacterial properties the basic polypeptide isolated by Bloom et al.² and the protamines and histones³ which are also basic linear polypeptides of relatively low molecular weight.

In view of the simple chemical structure of the synthetic poly-amino acids and of the similarity, in some measure, of their antibacterial properties to those of natural antibiotic peptides, it appears possible that the elucidation of the mechanism of action of water-soluble poly- α -amino acids on bacteria may shed some light on the mechanism of action of natural peptide antibiotics.

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THE INACTIVATION OF ESTRONE BY LIVER HOMOGENATES DETERMINED BY FLUOROMETRY

In 1934 B. Zondek^{1,2} found that estrone is inactivated by animals and plants. The discovery that the liver *in vitro* is capable of inactivating estrone^{1,2} pointed to the important role of this organ in the metabolism of estrogens. It could be shown that estrone incubated with liver pulp loses its biological potency. Heated liver pulp failed to effect the inactivation. It was concluded that the inactivation was enzymatic and the enzyme was called estrinase. In some experiments Zondek obtained cell-free extracts of liver with full enzymatic activity, but the results varied to a great extent. On the other hand, the plant enzyme could be extracted with water without difficulty^{1,2} and it has been shown that it is closely associated with tyrosinase and inactivates estrone, presumably by oxidation^{3,4,5}.

The nature of the liver enzyme involved in inactivation of estrone is still un-

known and we are not familiar with the mechanism of this inactivation. The difficulty in the study of this problem arises from lack of a suitable quantitative method for determination of the various estrogen metabolites and from the extreme lability of the enzyme(s) involved.

It has been the object of the present study:

- (1) to develop a convenient method for the determination of the natural estrogens in the presence of liver pulp, homogenate or extract, and
- (2) to establish the conditions for the preparation of a cell-free extract, capable of inactivating estrone; and further to purify the enzyme(s) concerned.

A method for the quantitative determination of natural estrogenic hormones in the presence of liver pulp (or homogenate or extract) has been elaborated. The extraction and purification of the estrogen fraction was accomplished by a modification—details of which will be given elsewhere—of the procedures of Mather⁶, and Bachman and Pettit⁷ for extraction of urinary estrogens.

The determination of the estrogens was accomplished fluorometrically^{8,9}. This procedure permits quantitative analysis of small amounts of estrogens used in the inactivation studies with the mean error of $\pm 15\%$. Table I shows representative

results of the determination of quantities of estrone unknown to the analyst added to liver pulp or homogenates to which immediately 5 volumes of ethanol were added to prevent inactivation.

TABLE I.

| Exp. No. | Estrone added | Estrone found | Recovery % |
|----------|---------------|---------------|------------|
| | γ | γ | |
| 1 | 10 | 8.7 | 87 |
| 2 | 7 | 5.5 | 78 |
| 3 | 11.3 | 8.0 | 70.7 |
| 4 | 14.0 | 15.0 | 107.1 |
| 5 | 10.0 | 9.0 | 90 |

We have found that liver pulp on incubation with estrone destroys the fluorescence exhibited by the hormone, and that the disappearance of fluorescence parallels the biological inactivation. Thus it has been convenient to measure the inactivation fluorometrically.

To obtain a cell-free active liver homogenate it has been found necessary to work at temperatures below 0°C . Homogenisation was carried out in the cold in a test tube, using a small motor-driven glass homogeniser. Addition of nicotinamide results in a more active preparation¹⁰.

Table II summarizes a few of the inactivation experiments. In each case 10 γ

TABLE II.

| Exp. No. | Liver preparation | Estrone added | Estrone found at the beginning of the Exp. (C_0) | Estrone found after 4 hrs. of incubation at 37°C (C_4). | Inactivation % |
|----------|-------------------|---------------|--|---|----------------|
| | | γ | γ | γ | |
| 1 | pulp | 10.0 | 7.5 | 3.0 | 60 |
| 1 | homogenate | 10.0 | 7.5 | 1.5 | 80 |
| 1 | " | 0.0 | 0.0 | 0.0 | 0 |
| 2 | " | 10.0 | 11.5 | 4.5 | 60 |
| 3 | " | 10.0 | 7.0 | 2.0 | 71.5 |
| 4 | pulp | 10.0 | 10.0 | 5.5 | 55 |
| 4 | " | 0.0 | 0.1 | 0.0 | 0 |

of estrone were added to the corresponding liver preparation in phosphate buffer of pH 7.1 containing 5% nicotinamide. An aliquot was removed immediately to determine fluorometrically the initial concentration of estrone (C_0). After four hours of incubation in a shaking incubator at 37°C., the estrone concentration was determined on another aliquot (C_4). From the difference between the concentrations (C_0-C_4) the percentage of inactivation, which was of the order of 55-80%, could be calculated.

These experiments show that cell-free homogenate of liver is able to inactivate estrone.

A detailed paper will be published elsewhere.

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THE EFFECT OF GONADOTROPIC HORMONE ON TUMOUR FORMATION IN INTRASPLENIC OVARIAN GRAFTS

Biskind and Biskind¹ succeeded in producing luteoma and granulosa cell tumours by complete ovariectomy and implantation of the whole or part of one ovary into the spleen of the same rat. Luteomas were present at least 160 days after the implantation, whereas granulosa cell tumours appeared not earlier than 300 days. No tumours were found when the implantation was made into the kidney.

Estrogen produced by the spleen transplants was presumably completely inactivated by the liver^{2,7} which received the entire blood flow of the spleen. The authors could not decide whether the disappearance of estrogen from the general circulation itself, or the resultant hypophyseal stimulation (due to failure of the usual inhibition of the hypophysis by the estrogenic hormone) was the more important factor in the production of these tumours.

If the hyperproduction of the anterior pituitary gonadotropin plays an important role, it should be possible to enhance the formation of luteomas and granulosa cell tumours by injecting the castrated rats chronically with gonadotropin after intrasplenic ovarian grafting.

In one experiment, twelve rats weighing from 110 to 170 g. were castrated in October, 1949, and on the same occasion a part of one of the ovaries was implanted into the spleen. About four weeks later, the animals were injected three times weekly with gonadotropin from 10 to 100 R.U.; three animals received pregnant mare blood gonadotropin (P.M.B.G.-Antex) in doses ranging from 10 to 100 R.U.; three animals received pregnant urine gonadotropin (P.U.G.—Korotrin) in doses ranging from 10 to 100 R.U.; four animals were injected with a mixture of both P.M.B.G. and

| No. Rat | Date of castration and transplantation | Injected with | Dosage | Injection started | Sacrificed on | Period from transplantation to sacrifice | Period of injection | Size of tumour | Findings in spleen |
|------------|---|---------------------|-----------------|----------------------|------------------|--|------------------------|-------------------|--|
| 1 | 140 g 26.10.49 | Antex | 10 R.U. | 27.11.49 | 8.3.50 | 133 days | 101 days | pinhead | Luteoma |
| 2 | 110 g 26.10.49 | " | 20 R.U. | " | 8.3.50 | 140 days | 101 days | | corp. lut. follicles |
| 3 | 160 g 26.10.49 | " | 100 R.U. | " | 8.3.50 | 120 days | 88 days | | foreign body granuloma |
| 4 | 160 g 26.10.49 | Korotrin | 10 R.U. | " | 8.3.50 | 127 days | 88 days | | corp. lut. follicles |
| 5 | 170 g 26.10.49 | " | 20 R.U. | " | 23.2.50 | 140 days | 101 days | 1x0.7 cm | Luteoma |
| 6 | 150 g 19.10.49 | " | 100 R.U. | " | 23.2.50 | 133 days | 101 days | | Granulosa cell carcinoma Luteoma |
| 7 | 130 g 19.10.49 | Antex & Korotrin | 10 & 10 R.U. | " | 23.2.50 | 146 days | 106 days | 6x7 mm | Luteoma |
| 8 | 130 g 19.10.49 | " | 20 & 20 R.U. | " | 13.3.50 | 120 days | 88 days | | corp. lut. follicles |
| 9 | 140 g 19.10.49 | " | 10 & 10 R.U. | " | 13.3.50 | 138 days | 106 days | | corp. lut. follicles |
| 10 | 150 g 18.10.49 | " | 10 & 10 R.U. | " | 14.3.50 | 146 days | 107 days | | corp. lut. follicles |
| 11 | 130 g 12.12.49 | — | — | — | 13.4.50 | 122 days | — | | cyst. foll. |
| 12 | 140 g 12.12.49 | — | — | — | 13.4.50 | 122 days | — | | corp. lut. follicles |

P.U.G.; and two animals served as un-injected controls.

Of the ten gonadotropin-treated animals, three developed luteomas (two, 101 days, and one, 106 days after beginning of injection), and one developed a granulosa cell tumour (101 days after beginning of injection). In five animals, only developing ovarian tissue was found (follicles and fresh corpora lutea); in one rat the implant did not take (see Table).

In this small series we observed no difference between the effect of P.M.B.G. and P.U.G. The only granulosa cell carcinoma (rat No. 5) was obtained with P.U.G.

In the two controls not treated with gonadotropin only ovarian tissue was found; there was no tumour formation.

A second experiment on twelve young rats was entirely negative. Castration was followed by ovarian implantation into one or both kidneys and the animals were subsequently treated with gonadotropins of various sources as above. No tumours were found and only normal functional ovarian tissue was present as evidenced by oestrus.

Tumours in animals with intrasplenic grafts appeared earlier (40-100 days) than in Biskind's experiments, in which the rats were not treated with gonadotropin. From this it may be concluded that injected gonadotropin is able to enhance the formation of ovarian tumours. The hyperproduction of gonadotropin after castration probably plays an important role in the stimulation of ovarian tumour formation as long as the estrogen produced by the graft disappears from the general circulation owing to destruction in the liver. (This investigation was aided by a grant from the Jane Coffin Childs Memorial Fund for Medical Research.)

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A SELECTIVE MEDIUM FOR THE ISOLATION OF *VIBRIO CHOLERAE*

The most extensively used media for the isolation of *Vibrio cholerae* are alkaline peptone water and Dieudonne's agar¹. They are, however, not always satisfactory, especially when *V. cholerae* has to be isolated from sources in which it is accompanied by Coliforms, spore formers, Cocci and Pseudomonas types. These micro-organisms are not completely inhibited by the alkalinity of the medium, and, when present in very large numbers, may impede the development of the *Vibrio*.

The purpose of the present investigation was to devise a medium selective for *V. cholerae*, which would also make possible its easy identification. The properties of *V. cholerae* on which this attempt was based were: its resistance to relatively high alkalinity^{1, 2} and salt concentration, and its ability to ferment saccharose and to hydrolyse starch. Eventually the following medium was developed:

| | |
|---------------|-------|
| Bacto-peptone | 0.5% |
| Meat extract | 0.6% |
| Starch | 1% |
| NaCl | 2% |
| Agar-Agar | 2-2½% |

After sterilizing at 120°C the following ingredients were added (per 100 cc. of the above):

- 1.5 cc sodium sulfite solution (10%)
- 4.0 cc sodium carbonate solution (20%)
- 2.5 cc saccharose solution (20%)
- 0.2 cc acid fuchsin, saturated alcoholic solution
- 1/200,000 gentian violet
- 1/200,000 brilliant green.

The final pH of the medium was 8.5-8.7.

In this medium, the colonies were round-edged, transparent and slightly pinkish. Their diameter was 2-3 mm, and they were surrounded by a clear zone, due to the decomposition of starch.

A fluid medium was prepared on the same lines, but without starch. After an incubation of 6 hours at 37°C, it showed the characteristic surface pellicle of *V. cholerae*.

The fluid medium inhibits the growth of Coliforms, spore formers and Pseudomonas for 12 hours, and of Cocci for 24 hours, whilst the pellicle of the *Vibrio cholerae* appears after the first 6 hours. In the solid medium, the accompanying organisms are inhibited for 48 hours, while the presence of *V. cholerae* can be detected after an incubation of 6-8 hours, by the appearance of the lytic zones around the would-be colonies.

In working with pure cultures, no differences in the number of colonies were found between the above solid medium and nutrient agar. When the solid medium was inoculated from faeces to which *V. cholerae* in a concentration of 10^{-1} per cc. had been added, all the accompanying organisms were inhibited, while *V. cholerae* gave colonies in accordance with its initial concentration.

In comparing the sensitivity of alkaline peptone water (pH 8.5-8.7) and the above fluid medium, it was found that *V. cholerae* could be isolated in the latter up to a dilution of 10^{-9} , while in the former the highest dilution to give positive results was 10^{-7} . In these experiments, too, faeces were used to which *V. cholerae* had been added.

The following working method is suggested. Inoculate 1 cc of the material to be tested, into 10 cc of the fluid medium. Incubate for 6 hours at 37°C. Transfer to the solid medium and incubate overnight. Stain pellicle and colony. Identify with specific antisera on slides.

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VARIATIONS OF VIRULENCE IN A STRAIN OF *PAST. TULARENSIS*

In an investigation of the biology of *Past. tularensis*, some observations were made which have a bearing on the variations in virulence which occur *in vitro* and *in vivo*.

(1) The variations on artificial media were investigated. The original strain was one of high virulence showing a LD_{50} of 10^{-10} for white mice. The strain was cultured on two different media, glucose-cystein-blood agar (g.c.b.a.), and the egg medium of McCoy and Chepin. After an incubation period of 24-48 hours, the slants were stored in a refrigerator at 4-6°C, and after 3 months the virulence of the strain was tested. During the same period of time, the strain was transferred fifty times on g.c.b.a. slants, and its virulence determined. The cultures stored in the refrigerator without transfer showed almost no change in virulence (LD_{50} $10^{-9.7}$) on either culture medium. In contrast to this, the strain transferred fifty times showed an almost thousandfold reduction in virulence (LD_{50} $10^{-7.3}$).

(2) A series of experiments was car-

ried out designed to solve the problem whether the virulence raised by passage through experimental animals was limited to the animal used for the passage, or applied also to other animals. For these experiments white mice and white rats were chosen, as these two species differ in their susceptibility for *Past. tularensis*. The original strain showed an LD₅₀ of 10^{-7.8} for mice and 10^{-1.8} for rats.

After six parallel passages through mice and rats, the virulence showed approximately a hundredfold increase (LD₅₀ 10⁻¹⁰ for mice; and 10^{-3.8} for rats). There was no difference in virulence for either animal between the strain that passed through mice and the one that passed through rats. The six passages were sufficient to raise the virulence for mice to the maximum, but the virulence for rats did not reach its maximum, as shown by the following experiments.

During 14 additional passages through mice, the virulence for rats did not change. There is, obviously, a limit in the increase of virulence for rats that can be obtained by passages through mice. Therefore, additional passages were made through rats. After 4 passages, the LD₅₀ reached 10^{-3.8}, and after 4 more passages, 10^{-4.6}.

These experiments show that the passages through the more susceptible animal (the mouse) bring about a maximum rise in virulence for that animal, and a considerable—but not maximum—rise for the less susceptible animal (the rat). On the other hand, passages through the rat increase the virulence for the mouse rapidly to the maximum, but in order to obtain maximum virulence for the rat, there is need of a considerable number of additional passages through that species.

(3) Some interesting facts came to light during an investigation of the behaviour of the micro-organism towards streptomycin. A streptomycin-resistant variant, when tested for its virulence, was found to be almost avirulent (LD₅₀ for mice 10⁻²).

Microscopic examination showed that the highly virulent strain had a capsule (indian ink strain), while the strain of low virulence had practically none. This suggests that the capsule is one of the factors determining the virulence of this micro-organism. The hypothesis is supported by the fact that a strain, which had become avirulent during transfers in the presence of an anti-serum, had also lost its capsule.

These findings conflict with the conclusions drawn by Hesselbrock and Foshay from their work on the morphology of *Past. tularensis*¹. They could not observe a true capsulation, nor detect any relation between the "thickness" of the wall and the virulence of the strain.

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EXPERIMENTS ON THE FORMATION OF THE PURINE NUCLEUS BY *E. COLI*

Shive¹ has found that sulfa-drugs have a very specific effect on *E. coli*: they cause an accumulation of 4-amino-imidazole-5-carboxamide (I). This effect is counteracted by p-aminobenzoic acid. It seems obvious that (I) is an intermediary in the biosynthesis of the purine skeleton (*e.g.* xanthine, [II]). Thus, the formation of (I) may indicate that there exists a fundamental difference in the process of the synthesis of purines by *E. coli* (and probably other bacteria) and by higher animals. In the latter, both C₂ and C₈ of the purine skeleton are derived from the same source (formate, or the β-carbon atom of serine [III]^{2, 3}), while in the former, the incorporation of C₈ is possible under conditions in which that of C₂ is inhibited.

Our experiments were designed to discover whether the source of the lone carbon atom at C_2 is the same for *E. coli* as for higher animals. It has now been shown that serine (III) does not decrease the production of (I) by sulfa-inhibited *E. coli*; it cannot, therefore, be the source of C_2 . Other possible sources of a single carbon atom are choline (IV), betaine (V), and methionine (VI)⁴. None of them counteracted the inhibiting effect of sulfadiazine on *E. coli*. When, however, methionine (VI) was administered together with catalytic quantities of p-aminobenzoic acid, the amount of (I) formed was reduced appreciably. As the combination of p-aminobenzoic acid with neither choline, betaine nor serine has any effect on the amount of (I) formed, the source of C_2 in the purine skeleton is likely to be methionine. On the other hand, one may tentatively assume that methionine is not the source of the carbon atom C_8 of the purine nucleus.

Some preliminary experiments have been made as to the source of C_8 ^{4a}. It has been found that both acetate and serine (III) increase to a certain extent the amount of (I) formed by *E. coli* (Fig. 1)

and this may be taken as an indication that the bacterium utilizes these two compounds for the synthesis of (I). However, there may be still other intermediates from which the bacterium can build up the purine skeleton, such as 4,5-diamino-2,6-dihydroxy-pyrimidine (VII); a systematic study of this problem is now being undertaken.

It is significant that methionine can supply the carbon atom C_2 only in the presence of p-aminobenzoic acid, and we may, therefore, suggest a possible mechanism

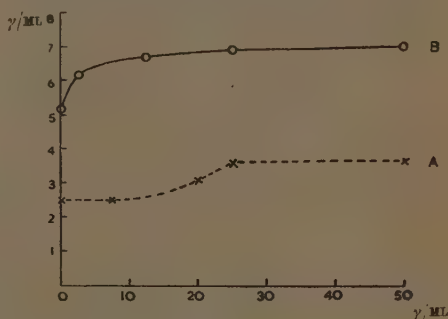
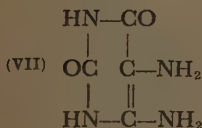
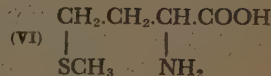
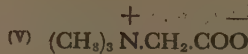
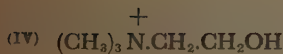
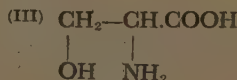
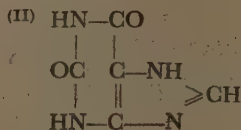
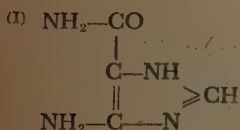


Fig. 1. — The effect of DL-serine (A) and acetic acid (B) on the accumulation of 4-amino-imidazole-5-carboxamide (I) by *E. coli*, in the presence of 0.5. mg.-% sulfadiazine.



ism of action of this amino-acid. *p*-Aminobenzoic acid may combine with the "labile methyl group"⁵ originating from methionine, forming an unstable compound, which can transfer the methyl group to (I). This suggests that the antagonistic effect of the sulfa-drugs is based on their higher affinity to that "labile methyl group"; they also combine with it, but the compounds formed are stable.

Mackenzie *et al.*⁶ and Sakami⁷ have assumed that the "labile methyl group" which is eliminated from the methionine molecule, is formic acid or a derivative of it. We have shown that neither formic acid nor a mixture of formic acid plus *p*-aminobenzoic acid can replace methionine as source of C₂: in neither case could there be observed a decrease in the amount of (I) produced by sulfa-inhibited *E. coli*. Moreover, N-formyl-*p*-aminobenzoic acid (VIII)⁸, which may be considered as a possible intermediate, showed activity only when applied in relatively high concentrations. It must be assumed, therefore, that another one-carbon compound is the active reagent in the conversion of (I) to (II).

A number of experiments have been carried out regarding the possible role of folic acid (pteroylglutamic acid) in the metabolism of *E. coli*. It could be assumed that *p*-aminobenzoic acid acts by virtue of its participation in the molecule of folic acid. However, it was found that folic acid does not counteract the sulfadiazine inhibition of bacterial growth, nor decrease the production of (I). This is in accord with the recent observations of Sevag, Koft and Steers⁹.

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THE ACTION OF CHEMOTHERAPEUTIC AND ANTIBIOTIC SUBSTANCES ON *LEPTOSPIRA GEFFEN*

The outbreak of Leptospirosis in Israel in 1948-49 induced a study of the epidemiology of the disease, its prevention and therapy.

It was found that the causative agent, *Leptospira Geffen*, is transmitted to humans by rodents (*Microtus guentheri*)¹. Albino Swiss mice infected with this *Leptospira* become carriers of the disease and constantly excrete the organism in their urine. This fact made it possible to study the effect of antibiotic and chemotherapeutic substances *in vivo*, in the same way as described by Meyer and Bruner² in their study of *Leptospira icterohaemorrhagiae* in hamsters.

The action of the following substances was studied:-

Dihydrostreptomycin sulfate-Pfizer, Aureomycin hydrochloride-Lederle, Chloramphenicol (chloromycetin)-Parke & Davis, Procain penicillin g-Squibb, Crystalline penicillin g-Glaxo, Sulfacetamide-Boots, Sulfathiazole, Sulfadiazine, Sulfanilamide-May & Baker, "Nitroakridine 3582"³.

In order to study their preventive action, these substances were injected into mice weighing 20-25 grams, either once, 24 hours, or three times, 24, 48 and 72 hours following the infection of each animal with 0.5 cc of a 5-7. days old Vor-

woort culture, which contained 20-30 living *Leptospira* per microscopic field. In the set of experiments designed to study the therapeutic action, the substances were injected into animals already excreting the *Leptospira* in their urine for a period of 7 to 30 days.

5 mg/ml of Chloromycetin did not prevent the appearance of the *Leptospira* in the urine even when administered three times, at intervals of 24 hours. Nor did it prevent the excretion of the *Leptospira* in mice which were carriers of the microorganism.

The same negative results were obtained with three injections of sulfathiazole (1 mg/ml), sulfadiazine (0.1 mg/ml), sulfacetamide (100 mg/ml), and sulfanilamide (15 mg/ml).

1 mg/ml of "Nitroakridine" did not prevent the excretion of *Leptospira* on the 5th day following infection, but proved lethal to mice 7 days following its administration.

Crystalline penicillin in a concentration of 2,000 units/ml prevents the excretion of *Leptospira*. But in animals which were already excreting the parasite in urine, no action was observed even following a course of six daily injections, for a period of four days, of 50,000 units per ml of crystalline penicillin, and one injection of 30,000 units of procain penicillin.

0.4 mg/ml of dihydrostreptomycin pre-

vents the appearance of *Leptospira*. But in order to destroy the microorganism already present in carriers, the concentration of the antibiotic had to be raised to 5 mg/ml. 0.3 mg/ml of dihydrostreptomycin inhibits the excretion of the *Leptospira* only for a period of 4 to 5 days, after which they reappear.

Aureomycin prevents the excretion of the microorganism in a concentration of 0.12 mg/ml. 0.1 mg/ml of aureomycin does not actually prevent, but postpones the excretion of the parasite. As in the case of dihydrostreptomycin, the excretion of the microorganism in the urine of carriers stops 48 hours following an injection of 0.36 mg/ml of aureomycin.

The results are summarized in the table below.

Conclusions:

1. The chemotherapeutic substances of the sulfonamide group and the "nitroakridine" proved ineffective against *Leptospira* Geffen as prophylactic and therapeutic agents, even in concentrations that approached the solubility limit of the sulfonamides and the toxicity limit of the "nitroakridine". Negative results were also obtained with chloromycetin.

2. Penicillin prevents the excretion of *Leptospira*, which are present in the circulating blood of the mouse¹, but is in-

| Substance | Therapeutic experiments | | Prophylactic experiments | |
|---------------------|--------------------------|-----------------------------------|---|-----------------------------------|
| | Concentration (mg/ml) | Excretion of <i>Leptospira</i> | Concentration (mg/ml) | Excretion of <i>Leptospira</i> |
| Sulfadiazine | 3x0.5 | + | 3x0.15 | + |
| Sulfathiazole | 3x1 | + | 3x1 | + |
| Sulfanilamide | 3x15 | + | 3x15 | + |
| Sulfacetamide | 3x100 | + | 3x100 | + |
| "Nitroakridine" | 1 | + | 3x1 | + |
| Chloromycetin | 3x5 | + | 3x5 | + |
| Dihydrostreptomycin | 0.4 | — | 5 | — |
| Aureomycin | 0.12 | — | 0.36 | — |
| Penicillin | 2,000(u/ml) | — | 50,000(u/ml)+ 30,000(u/ml) procain penicillin | + |

effective once the microorganism has disappeared from the blood stream and has localized itself in the internal organs of the animal.

3: Dihydrostreptomycin and aureomycin prevent, in the specified concentrations, the excretion of *Leptospira*, and free the carrier of the parasite. Lower concentrations do not prevent the recurrence of the disease. A similar effect of chloromycetin and aureomycin is known in cases of intestinal diseases.

Dihydrostreptomycin and aureomycin which are effective against *Leptospira* present in the blood, and also in the internal organs of the animal, are of some therapeutic value. The therapy in *Leptospirosis* begins usually at the period in which the parasite has already disappeared from the blood stream, and is found in the internal organs of the patient. At this stage, penicillin has no effect on the parasite.

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THE VALUE OF THE FEMALE GENITAL APERTURE FOR SPECIFIC DIAGNOSIS IN THE GENERA *AMBLYOMMA* AND *DERMACENTOR*

Adler and Feldman-Muhsam^{1,2} have shown that the female genital aperture was constant in Palestinian species of the genus *Hyalomma* and was therefore a valuable diagnostic character. This finding was of considerable practical importance since specific diagnosis on the basis of

other characters used by Koch³, Neuman⁴, Schulze⁵ and Senevet⁶ was, as Delpy⁷ pointed out, almost impossible in *Hyalomma*.

Although the genera *Dermacentor* and *Amblyomma* do not present the same diagnostic difficulties as did *Hyalomma* prior to the above finding, it is interesting to note that the female genital aperture differs significantly in various species of the above genera. (Determined species of these genera were kindly supplied by Dr. Glen M. Kohls from the Rocky Mountain Laboratory, U.S.A.).

In *Amblyomma*, the differences between three species studied, *A. cayennense*, *A. maculatum* and *A. americanum*, can be seen in both whole and mounted specimens. In *Dermacentor* differences appear clearly in mounted specimens, while in unmounted specimens only two groups can be distinguished among the species studied. The diagrams below are self-explanatory.

It will be interesting to determine whether the character of the female genital aperture is of taxonomic value in other genera of *Ixodidae*.

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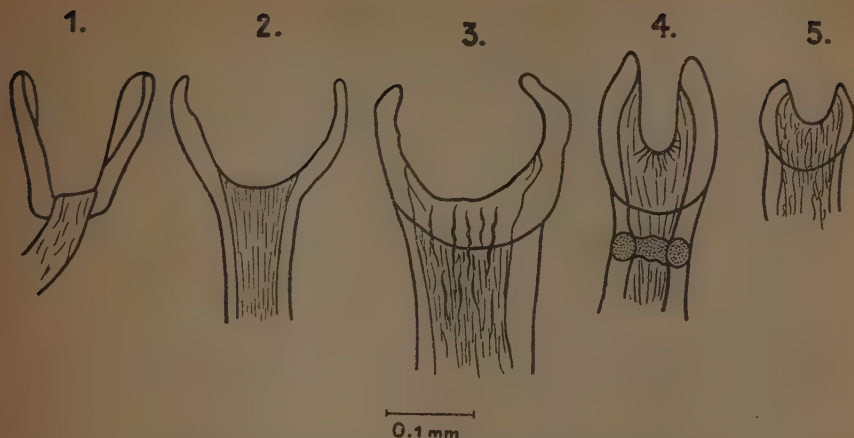


FIG. 1. The female genital aperture of mounted specimens of *Dermacentor*: 1. *D. occidentalis*; 2. *D. variabilis*; 3. *D. albipictus*; 4. *D. andersoni*; 5. *D. parumapertus*.

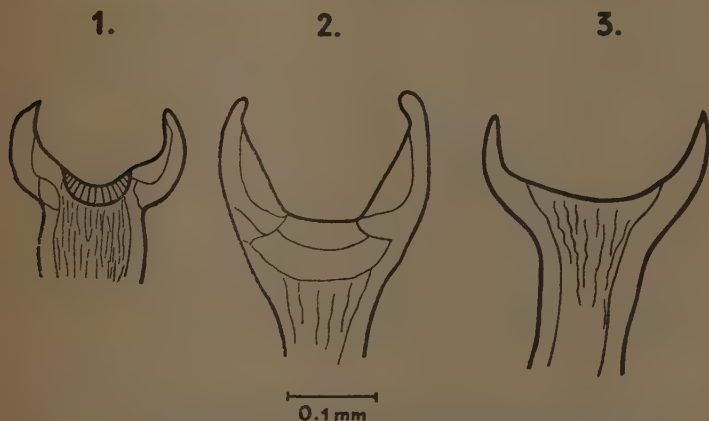


FIG. 2. The female genital aperture of mounted specimens of *Amblyomma*: 1. *A. americanum*; 2. *A. cayennense*; 3. *A. maculatum*.

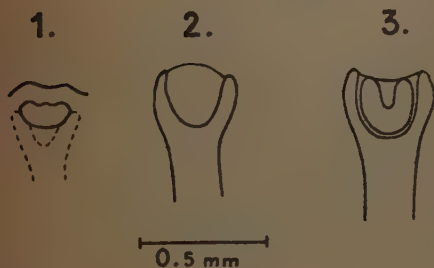


FIG. 3. The female genital aperture of unmounted specimens of *Amblyomma*: 1. *A. americanum*; 2. *A. cayennense*; 3. *A. maculatum*.

PHOXINELLUS (PARARHODEUS)
KERVILLEI PELLEGRIN FROM
LAKE HULEH, PALESTINE

The original description of this species by Pellegrin¹ is based upon specimens from the River Orontes in the Lebanon. The largest of his fishes were 44 mm total length. Tortonese³ added some details of specimens from Lake Huleh, collected by Festa in 1893.

Our specimens from Lake Huleh differ slightly from those of Festa-Tortonese. The following is a short account of these differences and of some further details not previously described.

Largest specimen: 58 mm total length, 48.5 mm standard length.

Height of body less than length of head. Although the index of standard-length to head, namely 3.5 — 3.7, was within the figures of Pellegrin (3.5—4.0), the length-height index (4.0—4.4) exceeded that of Pellegrin (3.5—4.0). The maximum head-eye index (3.6; specimen of 44 mm standard length) was larger than that given by Pellegrin. The interorbital space always exceeded the eye diameter (a character already mentioned by Tortonese for the "more developed" specimens), the proportion being 1.1—1.2. The pectoral is shorter than indicated by Pellegrin, the head-pectoral index being 1.8. The caudal peduncle is of much greater length than height, the ratio being 1.7—1.9.

The arrangement of the scales rows is not sufficiently regular to allow an accurate determination of the number of scales in the longitudinal line (37—42). The numbers of scales on the right and the left are rarely equal. Similarly, the incomplete lateral line frequently covers different numbers of scales on either side of the body (the smallest number is 8, the largest 18). Within the lateral-line row, one or two scales are sometimes not pierced by canals.

Dorsal fin: 7—8 branched rays preceded by (1) one soft, unbranched (simple) ray of nearly same length, (2) one

ossified spine considerably smaller than soft rays, (3) another ossified very small spine, according to usage not included in fin formula :2/7-8. Anal fin 2/7-8. Pectoral 1/13. Length of pectoral and of ventral more variable than indicated by Pellegrin or Tortonese: pectoral short of ventral or touching base of latter; ventral terminating before or behind anus, even reaching anal fin.

A dark longitudinal line is invariably present.

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THE INFLUENCE OF THE WATER CONTENT OF THE MEDIUM ON THE ACCUMULATION OF LARVAE AND PUPAE OF THE HOUSE-FLY

In experiments in breeding houseflies, it was observed that larvae reared in a mixture of bran, straw and water accumulated before pupation on the high side (drier part) of a container which stood in an inclined position (for the sake of convenience in the collection of pupae). The purpose of this investigation was to examine this phenomenon more thoroughly.

In three containers A, B, and C (140 x 50 x 20 cm each) containing 4½ kg of a 1:1 mixture of straw and bran and 12, 15 and 18 litres of water, respectively, 3 cc of eggs were spread on the surface of the medium. The containers were maintained at 28°C, no attempt being made to avoid the natural evaporation of water, and

were inclined lengthwise in such a way that the difference between the high and low side was 5 cm. After six days, when the larvae pupated, each medium was divided into six equal cross sections, each section taken out separately, and the pu-

pae collected and weighed. These sections were numbered consecutively, number 1 being the highest (driest) section and 6 the lowest (most humid). Results of a typical experiment are summarized in Table I.

TABLE I.

| Section of media | Container A (12 litres of water) | | Container B. (15 litres of water) | | Container C (18 litres of water) | |
|------------------|-------------------------------------|---------------------|--------------------------------------|---------------------|-------------------------------------|---------------------|
| | Weight of pupae (gr.) | Pupae in % of total | Weight of pupae (gr.) | Pupae in % of total | Weight of pupae (gr.) | Pupae in % of total |
| 1. | 115 | 56.3 | 173 | 50 | 193 | 58.1 |
| 2. | 38 | 18.6 | 124 | 35.8 | 125 | 37.6 |
| 3. | 29 | 14.2 | 41 | 11.8 | 14 | 4.2 |
| 4. | 17 | 8.3 | 8 | 2.3 | 0 | 0 |
| 5. | 5 | 2.4 | 0 | 0 | 0 | 0 |
| 6. | 0 | 0 | 0 | 0 | 0 | 0 |

It can be seen that the pupae tend to accumulate in the driest parts of the medium.

Similar experiments have shown that at 28°C the migration of larvae to the drier sections of the medium starts at the age of three days.

In order to determine the optimum humidity for the pupation of the larvae, a container, as described above, was divided by screens having openings of 0.5 cm², into six equal cross-sections, and these were provided with the following media of increasing water content:

| | | | % humidity |
|----|--------|----------------|---------------------|
| 1) | 760 gr | straw and bran | 0 |
| 2) | 760 | " " " " | + 600 cc water 44.1 |
| 3) | 760 | " " " " | +1,200 " " 61.2 |
| 4) | 760 | " " " " | +1,800 " " 70.3 |
| 5) | 760 | " " " " | +2,400 " " 76.0 |
| 6) | 760 | " " " " | +3,000 " " 80.0 |

The larvae could easily pass from one section to the next. After six days, when the larvae had pupated, each section of the medium was weighed to determine

the residual humidity, and the pupae from each section were collected and weighed. Results of one of the experiments are reproduced in Table II (temperature 28°C.)

TABLE II.

| Section of media | Weight of pupae (gr.) | Pupae in % of total | Weight of medium without pupae (gr.) | Final humidity (%) |
|------------------|-----------------------|---------------------|--------------------------------------|--------------------|
| 1 (driest) | 30 | 11.1 | 803 | 5.3 |
| 2 | 207 | 76.6 | 1,032 | 26.3 |
| 3 | 33 | 12.2 | 1,747 | 56.5 |
| 4 | 0 | 0 | 1,796 | |
| 5 | 0 | 0 | 1,910 | |
| 6 (most humid) | 0 | 0 | 2,106 | |

The larvae preferred to pupate in the section which initially contained 44% humidity; this humidity reached about 26% at the time of pupation. It was noted that the pupae which were found in the first (driest) section had concentrated at the

border of the second section, from which some water had been absorbed.

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